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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>6</sup> : <b>C09J 133/08, 121/00</b>	<b>A1</b>	(11) International Publication Number: <b>WO 96/25469</b> (43) International Publication Date: 22 August 1996 (22.08.96)
<p>(21) International Application Number: <b>PCT/US96/00430</b></p> <p>(22) International Filing Date: 11 January 1996 (11.01.96)</p> <p>(30) Priority Data: 08/390,780 16 February 1995 (16.02.95) US 08/578,010 22 December 1995 (22.12.95) US</p> <p>(71) Applicant: MINNESOTA MINING AND MANUFACTURING COMPANY [US/US]; 3M Center, P.O. Box 33427, Saint Paul, MN 55133-3427 (US).</p> <p>(72) Inventors: HYDE, Patrick, D.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). KRUEGER, Dennis, L.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). LAU, Felix, P.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). PYUN, Eumi; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). TUCKER, Pamela S.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US).</p> <p>(74) Agents: TRUESDALE, Carole et al.; Minnesota Mining and Manufacturing Company, Office of Intellectual Property Counsel, P.O. Box 33427, Saint Paul, MN 55133-3427 (US).</p>		<p>(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AZ, BY, KZ, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).</p> <p><b>Published</b> <i>With international search report.</i></p>
<p>(54) Title: <b>PRESSURE-SENSITIVE ADHESIVE</b></p> <p>(57) Abstract</p> <p>The present invention relates to pressure-sensitive adhesive compositions. The compositions are a blend of about 5 to 95 weight percent of an acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of a thermoplastic elastomeric copolymer. The compositions have a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to a major surface of the adhesive composition within said first domain. The invention further provides pressure-sensitive adhesive tapes and processes for preparing the compositions and tapes.</p> 		

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## PRESSURE-SENSITIVE ADHESIVE

### FIELD OF THE INVENTION

This invention relates to pressure-sensitive adhesive compositions, and, more particularly, pressure-sensitive adhesive compositions formed from at least two polymeric materials at least one of which is a pressure-sensitive adhesive, and to methods for making pressure-sensitive adhesives and articles having pressure-sensitive adhesives components.

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### BACKGROUND OF THE INVENTION

Pressure-sensitive adhesive tapes have been used for more than half a century for a variety of marking, holding, protecting, sealing and masking purposes. Pressure-sensitive adhesive tapes comprise a backing, or substrate, and a pressure-sensitive adhesive. Pressure-sensitive adhesives are materials which adhere with no more than applied finger pressure and are aggressively and permanently tacky. Pressure-sensitive adhesives require no activation other than the finger pressure, exert a strong holding force and should be removable from a smooth surface without leaving a residue.

Some of the earliest applications of adhesives in the medical field, where the product was referred to as an adhesive plaster, were not pressure-sensitive adhesives. These were, in fact, crude mixtures of natural rubber plasticized and tackified with wood resin derivatives and turpentine and heavily pigmented with zinc oxide. These tape-like products served their purpose but, with the advent of the truly pressure-sensitive adhesives they were replaced.

25

In the medical field, pressure-sensitive adhesive tapes are used for many different applications in the hospital and health areas, but basically they perform one of two functions. They are used to pull something in place, restricting movement such as in various strapping applications, or they are used to hold something in place, such as a wound dressing. It is important in each function that

the pressure-sensitive adhesive tape be compliant with and non-irritating to the skin and adhere well to the skin without causing skin damage on removal.

Similarly, early electrical tapes were black friction tape which is not pressure-sensitive; the adhesive is very soft and it splits when unwound. With the  
5 development of truly pressure-sensitive adhesives, these were also replaced. Generally, more current electrical tape has a pressure-sensitive adhesive applied to a plasticized polyvinyl chloride backing or a polyethylene or rubber film backing. Electrical tape is used to insulate, hold, reinforce and protect electrical wires. Other uses include providing a matrix for varnish impregnation, identifying wires  
10 in electrical circuitry, and protecting terminals during manufacture of electrical circuit boards. With electrical tape, it is important that the tape be stretchable, conformable and meet nonflammability requirements.

Packaging applications require a large variety of tapes for uses such as closing packages, protecting labels, sealing packages from moisture, and strapping  
15 and bundling loose parts. Packaging tapes are subjected to continuous shear and low angle peel forces. Generally, if the adhesive mass is of low cohesive strength, it fails in shear; if the shear resistance is improved by adding firmness to the adhesive, it has tendency to be less tacky and fail adhesively.

Pressure-sensitive adhesives require a delicate balance of viscous and  
20 elastic properties which result in a four-fold balance of adhesion, cohesion, stretchiness and elasticity. Pressure-sensitive adhesives generally comprise elastomers which are either inherently tacky, or elastomers or thermoplastic elastomers which are tackified with the addition of tackifying resins. They can be coated in solvent or as water-based emulsions to reduce the material viscosity to a  
25 level that is easily applied to a substrate of choice.

Major classes of pressure-sensitive adhesives include tackified natural rubbers; synthetic rubbers such as butyl rubber; and tackified linear, radial, star, branched and tapered block copolymers such as styrene-butadiene, styrene-ethylene/butylene and styrene-isoprene; polyvinyl ethers; acrylics, especially those  
30 having long chain alkyl groups; poly- $\alpha$ -olefins; and silicones.

Generally, when additives are used to enhance properties of pressure-sensitive adhesives they are required to be miscible with the pressure-sensitive adhesive or to have some common blocks or groups to permit homogeneous blends to form at the molecular level. Pressure-sensitive adhesives have been  
5 modified to extend their applicability into new areas. Tackified thermoplastic elastomers have been dissolved in acrylic monomers and subsequently cured. Tackified thermoplastic elastomers have also been added to polymerized acrylic pressure-sensitive adhesives in solvent where each component contains a common segment to permit compatibility. The general purpose is to combine the high shear  
10 properties of elastomers with the high tack performance of acrylics. Further improvements and better balance of properties continue to be sought.

### SUMMARY OF THE INVENTION

The present invention provides a pressure-sensitive adhesive composition  
15 comprising a blend of about 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of at least one thermoplastic elastomeric material, said composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature  
20 parallel to the major surface of the adhesive composition within said first domain. The thermoplastic elastomeric material may optionally contain a tackifying resin or plasticizer, in which case it also may be an adhesive.

The present invention further provides a process for preparing a pressure-sensitive adhesive composition comprising the steps of

- 25       (1)     melt blending about 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of at least one thermoplastic elastomeric material,
- (2)     (a)   forming said melt blended materials under shear and extensional conditions or



- (b) forming and drawing said melt blend  
to form a pressure-sensitive composition having a morphology comprising  
at least two distinct domains, a first domain being substantially continuous  
5 in nature and said second domain being fibrillose to schistose in nature  
parallel to the major surface of the adhesive within said first domain, and  
(3) allowing said composition to cool.

The present invention further provides a pressure-sensitive adhesive tape  
comprising a substrate with a pressure-sensitive adhesive composition comprising  
10 a blend of about 5 to 95 weight percent of acrylic pressure-sensitive adhesive and  
about 5 to 95 weight percent of thermoplastic elastomeric material, said  
composition having a morphology comprising at least two distinct domains, a first  
domain being substantially continuous in nature and said second domain being  
fibrillose to schistose in nature parallel to the major surface of the adhesive within  
15 said first domain on the face of the substrate.

The present invention also provides an electrical pressure-sensitive  
adhesive tape comprising a substrate and on the substrate a pressure-sensitive  
adhesive composition comprising a blend of about 5 to 95 weight percent of  
acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of  
20 thermoplastic elastomeric material, said composition having a morphology  
comprising at least two distinct domains, a first domain being substantially  
continuous in nature and said second domain being fibrillose to schistose in nature  
parallel to the major surface of the adhesive within said first domain. The  
substrate is preferably a polyvinyl chloride film or a film of a blend of ethylene-  
25 vinyl acetate and ethylene-propylene-diene rubber.

The present invention also provides a process for preparing a pressure-  
sensitive adhesive tape comprising the steps of

- (1) melt blending about 5 to 95 weight percent of at least one acrylic pressure-  
sensitive adhesive and about 5 to 95 weight percent of at least one  
30 thermoplastic elastomeric material,

(2) (a) forming said melt blended materials under shear or extensional conditions or both or

(b) forming and drawing said melt blend,

said blend forming an adhesive composition and said composition being extruded onto a face of a substrate to form a pressure-sensitive adhesive tape, said adhesive having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and

(3) allowing said adhesive to cool.

The present invention also provides a process for preparing a pressure-sensitive adhesive tape comprising the steps of

(1) melt blending about 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of at least one thermoplastic elastomeric material,

(2) (a) forming said melt blended materials under shear or extensional conditions or both or

(b) forming and drawing said melt blend to form a pressure-sensitive adhesive composition and

(c) coextruding a film forming polymeric resin with said adhesive composition,

said adhesive composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and

(3) allowing said composition and said polymeric resin to cool.

The present invention also provides a double coated pressure-sensitive adhesive tape comprising a substrate having on a first face a pressure-sensitive adhesive composition comprising a blend of about 5 to 95 weight percent of an acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of a

thermoplastic elastomeric material, said composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to the major surface of the adhesive within said first domain and having on  
5 a second face a pressure-sensitive adhesive which may be the same or different from that on the first face.

The present invention still further provides a process for forming a double coated pressure-sensitive adhesive tape comprising the steps of

10 (1) melt blending about 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of a thermoplastic elastomeric material,

(2) (a) forming said melt blended materials under shear or extensional conditions or both or

15 (b) forming and drawing said melt blend, to form a pressure-sensitive adhesive composition,

said composition being fed to the outer portions of a feed block having at least three layers and a film forming polymeric resin being fed to a middle portion of said feed block to form a double-coated pressure-sensitive adhesive tape, said adhesive composition having a morphology comprising  
20 at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and

(3) allowing said tape to cool.

The present invention also provides a pressure-sensitive adhesive  
25 composition for adhesion to skin comprising a substrate and on the substrate a pressure-sensitive adhesive composition comprising a blend of about 5 to 95 weight percent of acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of thermoplastic elastomeric material, said composition having a morphology comprising at least two distinct domains, a first domain being  
30 substantially continuous in nature and said second domain being fibrillose to



schistose in nature parallel to the major surface of the adhesive within said first domain. Suitable substrates, or backings include occlusive, i.e., substantially non-breathable, backings such as, for example, films, foam materials and laminates thereof, and non-occlusive, i.e., breathable, backings such as, for example,  
5 perforated polymeric film, woven fabrics, nonwoven fabrics such as hydroentangled or melt blown webs and thermally embossed nonwoven webs.

Increased peel adhesion over the acrylic pressure-sensitive adhesive alone and solution blended acrylic/thermoplastic elastomeric pressure-sensitive adhesives can be observed when the continuous domain is a pressure-sensitive adhesive and  
10 the fibrillose to schistose, i.e., discontinuous or co-continuous domain has elastic properties but is not pressure-sensitive. Controlled tack and peel adhesion properties can be observed when the continuous domain is not a pressure-sensitive adhesive. When each of the polymeric materials is a pressure-sensitive adhesive, the composition exhibits enhanced peel adhesion over similar blends of pressure-  
15 sensitive adhesives formed and coated from solvent.

The pressure-sensitive adhesives of the present invention are useful in such applications as in medical tapes; in box sealing tape; in electrical tape; and in repositionable tapes, removable tapes, and sheet materials. By proper selection of the polymeric materials, a variety of desirable end use properties can be designed  
20 into the adhesive. When used as adhesives for contact with skin, the compositions of the invention have enhanced initial adhesion over acrylic pressure-sensitive adhesives or the thermoplastic elastomeric material. Further, the aged adhesion to skin can be manipulated by, for example, adjusting the relative amounts of the acrylic pressure-sensitive adhesive and thermoplastic elastomer components.

25

### **BRIEF DESCRIPTION OF THE DRAWINGS**

FIG. 1 is a cross-sectional view of the adhesive layer of Example 5 taken along the film forming direction at 1000X using scanning electron microscopy (SEM).

FIG. 2 is a surface view of the adhesive layer of Comparative Example C6 at 10X using optical microscopy.

FIG. 3 is a cross-sectional view of the adhesive layer of Example 7 taken along the film forming direction at 6000X using transmission electron microscopy (TEM).

FIG. 4 is a cross-sectional view of the adhesive layer of Example 8 taken along the film forming direction at 6000X using TEM.

FIG. 5 is a cross-sectional view of the adhesive layer of Example 9 taken along the film forming direction at 6000X using TEM.

FIG. 6 is a cross-sectional view of the adhesive layer of Example 11 taken along the film forming direction at 3000X using SEM.

FIG. 7 is a cross-sectional view of the adhesive layer of Example 13 taken along the film forming direction at 2000X using SEM.

FIG. 8 is a cross-sectional view of the adhesive layer of Example 13 taken along the cross web direction at 2000X using SEM.

FIG. 9 is a cross-sectional view of the adhesive layer of Example 18 taken along the film forming direction at 1000X using SEM.

FIG. 10 is a cross-sectional view of the adhesive layer of Example 18 taken along the cross web direction at 1000X using SEM.

FIG. 11 is a cross-sectional view of the adhesive layer of Example 22 taken along the film forming direction at 2000X using SEM.

FIG. 12 is a cross-sectional view of the adhesive layer of Example 22 taken along the cross web direction at 2000X using SEM.

FIG. 13 is a cross-sectional view of the adhesive layer of Example 40 taken along the film forming direction at 1000X using SEM.

FIG. 14 is a plot of the dynamic mechanical analysis of Examples 24 - 26.

### **DETAILED DESCRIPTION OF THE INVENTION**

Acrylic pressure-sensitive adhesives generally have a glass transition temperature of about -20°C or less and may comprise from 100 to 80 weight

percent of a C<sub>3</sub>-C<sub>12</sub> alkyl ester component such as, for example, isooctyl acrylate, 2-ethyl-hexyl acrylate and n-butyl acrylate and from 0 to 20 weight percent of a polar component such as, for example, acrylic acid, methacrylic acid, ethylene vinyl acetate, N-vinyl pyrrolidone and styrene macromer. Preferably, the acrylic pressure-sensitive adhesives comprise from 0 to 20 weight percent of acrylic acid and from 100 to 80 weight percent of isooctyl acrylate. The acrylic pressure-sensitive adhesives may be self tacky or tackified. Useful tackifying resins for acrylics are rosin esters such as FORAL™ 85, available from Hercules, Inc., aromatic resins such as PICCOTEX™ LC-55WK, available from Hercules, Inc.,  
10 aliphatic resins such as ESCOREZ™ 1310LC, available from Exxon Chemical Co.

The second component of the pressure-sensitive adhesive compositions of the present invention is a thermoplastic elastomeric material which is blended with the acrylic pressure-sensitive adhesive. Thermoplastic elastomeric materials are materials which contain at least two segments, i.e., a hard segment and a soft  
15 segment, have a glass transition temperature greater than 50°C and exhibit elastic properties in one of the segments. The material is selected such that it is sufficiently incompatible and nonreactive with the pressure-sensitive adhesive at the use temperature to result in the pressure-sensitive adhesive composition having at least two distinct domains. Of course, more than one second component may  
20 be combined with the pressure-sensitive adhesive. The second component may or may not also be a pressure-sensitive adhesive.

Thermoplastic elastomeric materials useful in the present invention include, for example, linear, radial, star and tapered styrene-isoprene block copolymers such as KRATON™ D1107P, available from Shell Chemical Co. and  
25 EUROPRENE™ SOL TE 9110, available from EniChem Elastomers Americas, Inc., linear styrene-(ethylene-butylene) block copolymers such as KRATON™ G1657, available from Shell Chemical Co., linear styrene-(ethylene-propylene) block copolymers such as KRATON™ G1657X, available from Shell Chemical Co., styrene-isoprene-styrene block copolymers such as KRATON™ D1119P,  
30 available from Shell Chemical Co., linear, radial, and star styrene-butadiene block

copolymers such as KRATON™ D1118X, available from Shell Chemical Co., EUROPRENE™ SOL TE 6205, available from EniChem Elastomers Americas, Inc., polyetheresters such as HYTREL™ G3548, available from DuPont Co., and poly- $\alpha$ -olefin-based thermoplastic elastomeric materials such as those represented  
5 by the formula  $-(CH_2 CHR)_x$  where R is an alkyl group containing 2 to 10 carbon atoms and poly- $\alpha$ -olefins based on metallocene catalysis such as ENGAGE™ EG8200, an ethylene/1-octene copolymer available from Dow Plastics Co.

These thermoplastic elastomeric materials can be modified with tackifying resins or plasticizers to lower their melt viscosity to facilitate the formation of fine  
10 dispersions, with the smallest phase dimension preferably less than about 20 microns when blended with the acrylic pressure-sensitive adhesive. Tackifying resins or plasticizers useful with the thermoplastic elastomeric materials are preferably miscible at the molecular level, i.e., soluble in, any or all of the polymeric segments of the thermoplastic elastomeric material. The tackifying  
15 resins or plasticizers may or may not be miscible with the acrylic pressure-sensitive adhesive. The tackifying resin, when present can generally comprises about 5 to 300 parts by weight, more typically up to about 200 parts by weight, based on 100 parts by weight of the thermoplastic elastomeric material.

The acrylic pressure-sensitive adhesive and the thermoplastic elastomeric  
20 material are blended and coated using melt extrusion techniques. Mixing can be done by any method that results in a substantially homogeneous distribution of the acrylic pressure-sensitive adhesive and the thermoplastic elastomeric material.

The blend of the acrylic pressure-sensitive adhesive and the thermoplastic elastomeric material is prepared by melt mixing the components in the molten or  
25 softened state using devices that provide dispersive mixing, distributive mixing, or a combination of dispersive and distributive mixing. Both batch and continuous methods of blending may be used. Examples of batch methods include Brabender™ or Banbury™ internal mixing, and roll milling. Examples of continuous methods include single screw extruding, twin screw extruding, disk  
30 extruding, reciprocating single screw extruding, and pin barrel single screw

extruding. The continuous methods can include both distributive elements such as cavity transfer elements such as CTM™, available from RAPRA Technology, Ltd., Shrewsbury, England, pin mixing elements, and static mixing elements and dispersive elements such as Maddock mixing elements or Saxton mixing elements.

5           After the mixing step, the softened or molten acrylic pressure-sensitive adhesive and thermoplastic elastomeric material blend is formed into coatings which have a morphology such that the pressure-sensitive adhesive forms a substantially continuous domain and the thermoplastic elastomeric material forms a domain which is fibrillose to schistose in nature by processes that involve either  
10   shear or extensional deformations or both. When a tackifying agent is blended with the thermoplastic elastomeric material such that this too is now an adhesive material, then either adhesive domain may be continuous or the domains may be co-continuous. These processes may be either batch or continuous.

          An example of a batch process is the placement of a portion of the blend  
15   between the desired substrate to be coated and a release liner, pressing this composite structure in a heated platen press with sufficient temperature and pressure to form a pressure-sensitive coating of the desired thickness and cooling the resulting coating.

          Continuous forming methods include drawing the pressure-sensitive  
20   adhesive composition out of a film die and subsequently contacting a moving plastic web or other suitable substrate. A related continuous method involves extruding the pressure-sensitive adhesive composition and a coextruded backing material from a film die and subsequently cooling to form a pressure-sensitive adhesive tape. Other continuous forming methods involve directly contacting the  
25   pressure-sensitive adhesive blend to a rapidly moving plastic web or other suitable substrate. In this method, the pressure-sensitive adhesive blend can be applied to the moving web using a die having flexible die lips such as a reverse orifice coating die. After forming, the pressure-sensitive adhesive coatings are solidified by quenching using both direct methods, such as chill rolls or water baths, and  
30   indirect methods, such as air or gas impingement.



Preferably, each of the polymeric components has similar melt viscosity. The ability to form a finely dispersed morphology is related to the viscosity ratio and concentration of the components. The shear viscosity is determined using capillary rheometry at a shear rate approximating extrusion blending conditions, i.e.,  $100\text{s}^{-1}$  and  $175^{\circ}\text{C}$ . When a higher viscosity polymeric material is present as the minor component, the viscosity ratio of the minor component to the major component is preferably less than about 20:1, more preferably less than about 10:1. When a lower viscosity polymeric material is present as the minor component, viscosity ratios of the minor component to the major component are preferably greater than about 1:10, more preferably greater than about 1:5. The melt viscosities of individual polymeric materials may be altered by the addition of plasticizers, tackifying resins or solvents or by varying mixing temperatures. If the use of solvent is required, the solvent is preferably removed before the coating step to prevent foaming.

It is also preferable that at least one of the polymeric materials be easily extended in the melt blending and coating operations to form a finely dispersed morphology with domains which are fibrillose to schistose, e.g., forming sheets, ribbons, fibers, ellipsoids or the like, oriented in the web formation direction in the substantially continuous or co-continuous domain of the other polymeric material. Sufficient interfacial adhesion between the acrylic pressure-sensitive adhesive component and the thermoplastic elastomeric component preferably exists to withstand the shear and extensional deformation present during the forming step and to promote formation of a continuous film.

If none of the polymeric materials can be easily extended in the melt blending and coating or sufficient interfacial adhesion is not present, a pressure-sensitive adhesive coating may be produced which has gross discontinuities and is grainy in texture. Through use of suitably selected conditions of mixing, closeness of melt viscosities, and shear/stretch conditions during extrusion, the thickness of the fibrillose to schistose domains can be made sufficiently thin that delamination from the substantially continuous or cocontinuous domain will not occur.

Preferably, the thickness of the fibrillose to schistose domains is less than about 20 microns, more preferably less than about 10 microns, and most preferably less than about 1 micron although the size will vary depending on specific blends, i.e., polymer types, concentration, viscosity, and the like.

5           This invention is further illustrated by the following examples which are not intended to limit the scope of the invention. In the examples, all parts, ratios and percentages are by weight unless otherwise indicated. The following test methods were used to evaluate and characterize polymeric materials and the pressure-sensitive adhesive compositions produced in the examples.

10

#### Shear Viscosity

          Shear viscosity was determined using a high pressure capillary rheometer (RHEOGRAPH 2001, available from Gottfert Co.) operated with a capillary die 30 mm long and 1 mm in diameter at a temperature of 175°C. At a 100s<sup>-1</sup> shear  
15   rate, the apparent viscosity was calculated from Poiseuille's equation and converted to true viscosity using the Weissenberg-Rabinovitch correction.

#### 180° Peel Adhesion Test

          Pressure-sensitive adhesive tape samples 1.25 cm wide and 15 cm long  
20   were tested for 180° peel adhesion to glass and/or smooth cast biaxially oriented polypropylene films. The samples were adhered to the test surfaces by rolling the tapes with a 2.1 Kg (4.5 lb.) roller using 4 passes. After aging at ambient temperatures (~22°C) for approximately 1 hour, the tapes were tested using a Model 3M90 slip/peel tester, available from Instrumentors, Inc., in 180° geometry  
25   at 30.5 cm/min (12 in/min) peel rate, unless otherwise noted.

#### Shear Strength Test

          Shear strength was measured on pressure-sensitive adhesive tape samples at ambient temperatures. A 12.7 mm x 12.7 mm (0.5 in x 0.5 in) section of the  
30   tape was adhered to a stainless steel sheet with a 2.1 Kg (4.5 lb.) roller using 4

passes. A 1000 gram weight was adhered to the sample. The amount of time for the weight to drop was recorded.

#### Rolling Ball Tack Test

- 5        The rolling ball tack test was performed according to Pressure-Sensitive Tape Council Test PSTC-6 wherein a 7/16 inch steel ball was rolled down an inclined ( $21^{\circ}30'$ ) runway onto a sample of tape placed at the bottom of the runway on a flat surface. The result was the distance the ball rolls on the tape before stopping.

10

#### Electrical Tape Adhesion Test

- Pressure-sensitive adhesive electrical tape samples were tested against steel panel (adhesion to steel) and against web material (adhesion to backing) at a speed of 30.5 cm/min according to the American Society for Testing and Materials (ASTM) Standard Methods of Testing Pressure-Sensitive Adhesive-Coated Tapes Used for Electrical Insulation, ASTM D1000-79, Procedure A.
- 15

#### Skin Adhesion Test

- Skin adhesion testing was carried out by placing tape samples 2.5 cm wide by 7.5 cm long on the back of a human subject. Each tape was rolled down with one forward and one reverse pass using a 2 Kg roller moved at a rate of about 30 cm/min. Adhesion to the skin was measured as the peel force required to remove the tape at  $180^{\circ}$  angle at a 15 cm/min rate of removal. Adhesion was measured immediately after initial application ( $T_0$ ) and after either 24 hours ( $T_{24}$ ) or 48 hours ( $T_{48}$ ). Preferred skin adhesion generally exhibits a  $T_0$  of between about 30 and 100 grams (1.2 to 3.8 N/dm), a  $T_{24}$  of between about 150 and 300 grams (5.8 to 11.5 N/dm), and a  $T_{48}$  of between about 150 and 300 grams (5.8 to 11.5 N/dm).
- 20
- 25

**Skin Adhesion Lift Test**

When either the 24 hour or 48 hour skin adhesion test was performed, the tape sample was examined for the amount of area that was lifted (released) from the skin prior to removal of the tape and ratings were given as:

- 5           0    no visible lift  
          1    lift only at edges of tape  
          2    lift over 1% to 25% of test area  
          3    lift over 25% to 50% of test area  
          4    lift over 50% to 75% of test area  
10          5    lift over 75% to 100% of test area

Results of 9 tests were averaged. Preferred skin adhesives will generally exhibit an average rating below about 2.5.

**Skin Adhesive Residue Test**

- 15           When either the 24 hour or 48 hour skin adhesion test was performed, the skin underlying the tape sample was visually inspected to determine the amount of adhesive residue on the skin surface and was rated as:

- 0    no visible residue  
          1    residue only at edges of tape  
20          2    residue covering 1% to 25% of test area  
          3    residue covering 25% to 50% of test area  
          4    residue covering 50% to 75% of test area  
          5    residue covering 75% to 100% of test area

- Results of 9 tests were averaged. Preferred skin adhesives will generally exhibit  
25   an average rating below about 2.5.

**Examples 1-6 and Comparative Example C1**

- In Example 1, an acrylic pressure-sensitive adhesive (95 weight percent isooctyl acrylate/5 weight percent acrylic acid, water emulsion polymerized, shear  
30   viscosity - 150 Pa-s), prepared according to U.S. Pat. No. RE 24,906, (Ulrich)

which is incorporated herein by reference, and dried, and thermoplastic elastomeric KRATON™ D1107P (styrene-isoprene-styrene block copolymer, shear viscosity 1250 Pa-s) which was preblended with 1 weight percent IRGANOX™ 1010 antioxidant, available from Ciba-Geigy Corp. were melt-  
5 blended in a 34 mm diameter fully intermeshing co-rotating twin-screw extruder (LEISTRITZ™ Model LSM34GL, available from Leistritz, Inc.). The thermoplastic elastomeric block copolymer was introduced into the feed throat of the extruder and the acrylic pressure-sensitive adhesive was introduced in zone 4. The temperature was progressively increased from 38°C to 177°C (100°F to  
10 350°F) from zone 1 to zone 4. The temperature of the remaining zones was maintained at 177°C to 191°C (350°F to 375°F). The feed rates were adjusted to provide a ratio of pressure-sensitive adhesive to thermoplastic elastomeric block copolymer of 75:25.

The twin-screw extruder was continuously discharged at a pressure of at  
15 least about 0.69 MPa (100 psi) into a three-layer feed block (CLOEREN™ Model 92-1033, available from The Cloeren Co.) mounted on a 25.4 cm (10 inch) wide film die (UTLTRAFLX™ 40 die, Model 89-12939, available from Extrusion Dies, Inc.). The die was maintained at 177°C to 191°C (350°F to 375°F) and the die gap was 0.5 to 0.8 mm (20 to 30 mils). The blended adhesive composition  
20 was extruded through the outer two portions of the feed block, with the center portion of the feed block not used, and was fed between a 51 µm (2 mil) thick biaxially oriented polyethylene terephthalate film and a release coated paper web at a rate of 6.4 Kg/hr (14 lbs/hr). The film and the web were fed at a rate of 7.3  
25 m/min (24 fpm) between chill rolls maintained at a temperature of 21°C (70°F) to form a pressure-sensitive adhesive tape. In Examples 2, 3 and 4, the thermoplastic elastomeric block copolymer, KRATON™ D1117P (styrene-isoprene-styrene block copolymer; shear viscosity 690 Pa-s) preblended with 1 weight percent IRGANOX™ 1010, was added to the acrylic pressure-sensitive adhesive at ratios  
30 of acrylic adhesive to thermoplastic elastomeric block copolymer of 75:25, 63:37, and 25:75, respectively. In Example 5, the thermoplastic elastomeric block



copolymer was KRATON™ D1118X (butadiene-styrene block copolymer; shear viscosity 1160 Pa-s), which was added to the acrylic pressure-sensitive adhesive at a ratio of acrylic adhesive to thermoplastic elastomeric block copolymer of 82:18. In Example 6, the thermoplastic elastomeric polymer was STEREON 827,  
 5 (styrene-butadiene multi-block copolymer available from Firestone Synthetic Rubber & Latex Co.; shear viscosity 2040 Pa-s) which was added to the acrylic adhesive at a ratio of acrylic adhesive to thermoplastic elastomeric block polymer of 82:18. In Comparative Example C1, only the acrylic pressure-sensitive adhesive, with no other polymer was used to prepare the pressure-sensitive  
 10 adhesive tape.

The viscosity ratio of the discontinuous to substantially continuous component and the thickness of adhesive on samples of each pressure-sensitive adhesive tape were determined and the 180° peel adhesion test on glass and the 180° peel adhesion test on biaxially oriented polypropylene (BOPP) were carried  
 15 out. The results are set forth in Table 1. The morphology of the adhesive compositions of Examples 1-6 was also determined.

Table I

Example	Viscosity Ratio	Thickness (μm)	180° peel adhesion (N/dm)	
			Glass	BOPP
1	8.3:1	36	60	26
2	4.6:1	41	78	23
3	4.6:1	41	64	25
4	1:4.6	46	36	20
C1	-	76	48	27
5	7.7:1	76	76	32
6	13.6:1	76	86	29

As can be seen from the data in Table 1, the addition of the thermoplastic  
 20 elastomeric block copolymers to the acrylic pressure-sensitive adhesive increased the peel adhesion of the adhesive to both glass and biaxially oriented polypropylene in Examples 1, 2, 3, 5, and 6. Example 4 demonstrates that the adhesive properties of the adhesive composition can be controlled when excessive thermoplastic elastomeric block copolymer is used. The peel values of this

adhesive indicate that it would be suitable as a repositionable adhesive. In the Examples, the acrylic adhesive formed a substantially continuous domain and the morphology was schistose or ribbon-like. In Example 4, the thermoplastic elastomeric block copolymer formed a substantially continuous domain and the morphology was schistose. A scanning electron microscope (SEM) photomicrograph of a cross-section of an osmium stained adhesive layer of Example 5 is shown in FIG. 1, with the acrylic adhesive the dark portion.

#### Examples 7-9 and Comparative Examples C2-C6

10 In Examples 7-9, pressure-sensitive adhesive tapes were prepared as Example 1 except a thermoplastic elastomeric pressure-sensitive adhesive prepared from a thermoplastic elastomer (50 parts by weight KRATON™ D1107P) with tackifying resin (50 parts by weight WINGTACK PLUS™, available from Goodyear Tire and Rubber Co.) and antioxidant (1 part by weight 15 IRGANOX™ 1010) was substituted for the thermoplastic elastomeric block copolymer. This adhesive had a shear viscosity of 35 Pa-s at 191°C (375°F). The ratio of acrylic pressure-sensitive adhesive to thermoplastic elastomeric pressure-sensitive adhesive was 70:30, 50:50 and 25:75, respectively, and the acrylic adhesive was added to the extruder at the feed throat and the thermoplastic 20 elastomeric adhesive was added in zone 4. The pressure-sensitive adhesive tape of Comparative Example C2 was prepared using only the acrylic pressure-sensitive adhesive with no other polymer. The pressure-sensitive adhesive tape of Comparative Example C3 was prepared using only the thermoplastic elastomeric block copolymer KRATON™ D1107P preblended with antioxidant, with 25 tackifying resin WINGTACK™ Plus at a 50/50 weight ratio.

The thickness of each adhesive was determined and the 180° peel adhesion to glass test and the 180° peel adhesion to biaxially oriented polypropylene (BOPP) test were carried out. The results are set forth in Table 2.

In Comparative Examples C4, C5, and C6, the acrylic pressure-sensitive 30 adhesive and the pressure-sensitive adhesive prepared with the block copolymer

KRATON™ D1107P and the tackifying resin WINGTACK™ PLUS used in Examples 6-8 were dissolved in tetrahydrofuran at ratios of acrylic adhesive to block copolymer/tackifying resin adhesive of 70:30, 50:50 and 25:75, respectively, with solutions containing 20 weight percent adhesives and 80 weight percent solvent. The solutions were coated on 76  $\mu\text{m}$  (3 mil) thick biaxially oriented polyethylene terephthalate film and dried at room temperature to obtain coating thicknesses of 38  $\mu\text{m}$ . The thus-prepared pressure-sensitive adhesive tapes were tested for 180° peel adhesion to glass. The results are set forth in Table 3.

10

Table 2

Example	Thickness ( $\mu\text{m}$ )	180° peel adhesion (N/dm)	
		Glass	BOPP
C2	48	51	26
7	43	64	39
8	48	81	43
9	64	75	49
C3	56	95	65

As can be seen from the data in Table 2, addition of the thermoplastic elastomeric pressure-sensitive adhesive to the acrylic pressure-sensitive adhesive increased both 180° peel adhesion to glass and biaxially oriented polypropylene.

15 With regard to the morphologies of these adhesive compositions, each of Examples 7-9 was schistose in nature, i.e., stratified. In Example 7, the acrylic adhesive formed a substantially continuous domain, while the thermoplastic elastomer adhesive was discontinuous. In Example 8, the domains were substantially co-continuous. In Example 9, the thermoplastic elastomer adhesive  
20 was substantially continuous, while the acrylic adhesive was discontinuous.

Table 3

Example	Peel Adhesion (N/dm)
C4	35
C5	33
C6	30

As can be seen from the data in Tables 2 and 3, the extrusion melt blended adhesives of the invention perform significantly better than the same adhesive compositions prepared by a solvent coating procedure. With regard to the morphologies of these solution coated adhesives, each had a continuous domain of acrylate adhesive in Comparative Examples C4 and C5 or thermoplastic elastomer adhesive in Comparative Example C6 with discontinuous spheres of thermoplastic elastomer adhesive in Comparative Examples C4 and C5 and discontinuous spheres of acrylic adhesive in Comparative Example C6.

#### Examples 10-11 and Comparative Example C7

Double coated pressure-sensitive adhesive tape was prepared in Examples 10 and 11 using the melt blending and extrusion procedures as in Example 1 except the weight ratio of acrylic pressure-sensitive adhesive to thermoplastic elastomeric polymer was 85:15 and 70:30, respectively, and a polypropylene melt was fed to the middle portion of the CLOERENT<sup>TM</sup> feed block. The melts were coextruded to produce a double-coated pressure-sensitive adhesive tape having a polypropylene core of about 76 to 127  $\mu\text{m}$  (3 to 5 mil) thickness with 25 and 38  $\mu\text{m}$  (1.0 and 1.5 mil) thick adhesive coatings. In Comparative Example C7, a double-coated pressure-sensitive adhesive tape was prepared as in Example 10 and 11 except the thermoplastic elastomer was omitted. Each double-coated pressure-sensitive tape was tested for 180° peel adhesion from glass. The results of Examples 10, and 11 and Comparative Example C7 were 67 N/dm (1730 g/in), 74 N/dm (1913 g/in) and 51 N/dm (1332 g/in), respectively. As can be seen, the addition of the thermoplastic elastomer to the acrylic adhesive increased the 180°

peel adhesion of the adhesive significantly. In each of Examples 10 and 11, the morphology was schistose with the acrylic adhesive forming a substantially continuous domain with the thermoplastic elastomer forming a ribbon-like discontinuous domain. FIG. 6 shows the adhesive of Example 11 in cross-section  
5 viewed in the film forming direction after staining with osmium tetroxide at 3000X.

#### Examples 12 and 13 and Comparative Example C8

In Examples 12 and 13, double-coated pressure-sensitive adhesive tapes  
10 were prepared as in Examples 10 and 11, respectively except for the replacement of the water emulsion polymerized acrylic pressure-sensitive adhesive with a suspension polymerized composition. The suspension polymerized acrylic pressure-sensitive adhesive was prepared in accordance with U.S. Pat. No. 4,833,179 (Young et al.) in the following manner: A two liter split reactor  
15 equipped with condenser, thermowell, nitrogen inlet, stainless steel motor-driven agitator, and a heating mantle with temperature control was charged with 750g deionized water, to which was added 2.5 g of zinc oxide and 0.75 g hydrophilic silica (CAB-O-SIL™ EH-5, available from Cabot Corp.) and was heated to 55°C while purging with nitrogen until the zinc oxide and silica were thoroughly  
20 dispersed. At this point, a charge of 480 g isooctyl acrylate, 20 g methacrylic acid, 2.5 g initiator (VAZO™ 64, available from DuPont Co.) and 0.5 g isooctyl thioglycolate chain transfer agent were mixed together. The resulting solution with initiator and chain transfer agent was then added to the initial aqueous mixture while vigorous agitation (700 rpm) was maintained to obtain a good  
25 suspension. The reaction was continued with nitrogen purging for at least 6 hours, during which time the reaction was monitored to maintain a reaction temperature of less than 70°C. The resulting pressure-sensitive adhesive was collected and machine pressed to at least 90% solids by weight. In Comparative Example C8, a double-coated pressure-sensitive adhesive tape was prepared as in  
30 Examples 12 and 13 except the thermoplastic elastomer adhesive was omitted.



Each tape was tested for 180° peel adhesion to glass. The results for Examples 12 and 13 and Comparative Example C8 were 32 N/dm (830 g/in), 74 N/dm (1913 g/in) and 24 N/dm (615 g/in), respectively, demonstrating that the addition of the thermoplastic elastomer, particularly at higher levels increases the peel strength of the acrylic adhesive.

In Examples 12 and 13, the acrylic adhesive domain was substantially continuous and the thermoplastic elastomer formed ribbon-like layers, i.e., was schistose in nature. FIG. 7 shows the adhesive of Example 13 in cross-section viewed in the film forming direction after staining with osmium tetroxide at 2000X. FIG. 8 shows the adhesive of Example 13 in cross-section viewed perpendicular to the film-forming direction after staining with osmium tetroxide at 2000X.

#### Examples 14-17 and Comparative Example C9

In Examples 14 and 15, double-coated pressure-sensitive adhesive tapes were prepared as in Examples 12 and 13, respectively, except the acrylic adhesive used was an acrylic pressure-sensitive adhesive prepared by water suspension polymerization of 465 g isooctyl acrylate, 20 g methacrylic acid, 15 g methacrylate-terminated polystyrene macromer (CHEMLINK™ 4500, available from Sartomer Co.) 0.25 g acryloxybenzophone and 0.35 g isooctyl thioglycolate. In Comparative Example C9, a double-coated pressure-sensitive adhesive tape was prepared as in Examples 14 and 15 except the thermoplastic elastomeric block copolymer KRATON™ D1107P was omitted. In Examples 16 and 17, double-coated pressure-sensitive adhesive tapes were prepared as in Examples 14 and 15, respectively, except the thermoplastic elastomeric block copolymer KRATON™ G1657 (styrene -(ethylene-butylene)-styrene) was substituted for the KRATON™ D1107P. The peel adhesion to glass was determined for each pressure-sensitive adhesive tape. The results are set forth in Table 4.

Table 4

Example	180° Peel Adhesion (N/dm)	Shear Strength (minutes)
C9	32	27
14	55	37
15	66	29
16	17	43
17	22	38

As can be seen from the data in Table 4, addition of the KRATON™ D1107P improved the 180° peel adhesion of the suspension polymerized acrylic adhesive, while addition of the KRATON™ G1657 improved the shear strength of the adhesive. The addition of the KRATON™ G1657 did not improve the 180° peel adhesion of the adhesive because the elastic properties of the thermoplastic elastomer were insufficient in this particular blend. In each of Examples 14 to 17, the acrylic adhesive domain was substantially continuous with the thermoplastic elastomer domain being schistose in Examples 14 and 15 and fibrillose in Examples 16 and 17.

#### Examples 18 and 19

In Example 18, the acrylic pressure-sensitive adhesive used in Example 1 was melt-blended with a thermoplastic elastomeric block copolymer KRATON™ D1320X, (a multi-arm star styrene/isoprene polymer available from Shell Chemical Co., which had been preblended with 1 weight percent IRGANOX™ 1010) at a ratio of acrylic adhesive to thermoplastic elastomeric block copolymer of 80:20 using a corotating twin screw extruder (Model ZSK 30, available from Werner & Pfleiderer Corp., Ramsey, NJ, having a 30 mm diameter barrel and a length to diameter ratio of 45:1) with the thermoplastic elastomeric block copolymer being fed into zone I and the acrylic pressure-sensitive adhesive being fed into zone 3. The blend was extruded onto polyethylene film using a contact die with rates similar to those used in Example 1 to form a pressure-sensitive adhesive

tape. In Example 19, a pressure-sensitive adhesive tape was prepared as in Example 24 except a polyolefin thermoplastic elastomer (ENGAGE™ EG8200, available from Dow Plastics Co.; shear viscosity 1030 Pa-s) was substituted for the thermoplastic elastomeric block copolymer and the ratio of acrylic adhesive to thermoplastic elastomer was 90:10. The adhesive thickness and 180° peel adhesion value from glass were determined and are set forth in Table 5.

Table 5

Example	Adhesive Thickness (μm)	Peel Adhesion (N/dm)
18	47	>77
19	36	63

As can be seen from the data in Table 5, the adhesive composition prepared from the acrylic pressure-sensitive adhesive and the thermoplastic elastomeric polymers had excellent 180° peel adhesion values. Each of Examples 18 and 19 had a substantially continuous acrylic adhesive domain. In Examples 18 and 19, the thermoplastic elastomers formed schistose and fibrillose domains, respectively. FIG. 9 shows the adhesive of Example 18 in cross-section in the film forming direction after staining with osmium tetroxide. FIG. 10 shows the adhesive of Example 18 in cross-section along the cross direction after staining with osmium tetroxide.

#### Examples 20-23 and Comparative Example C10

In Example 20, 75 parts by weight of the acrylic pressure-sensitive adhesive used in Example 1, and a blend of 12.5 parts by weight thermoplastic elastomeric block copolymer KRATON™ D1107P, 1 part by weight antioxidant IRGANOX™ 1010 and 12.5 parts by weight tackifying resin ESCOREZ™ 1310LC, available from Exxon Chemical Co., were melt-blended and extruded as in Example 18 to form a pressure-sensitive adhesive tape. In Example 21, a

pressure-sensitive adhesive tape was prepared as in Example 20 except the proportions were 50 parts by weight acrylic adhesive, 25 parts by weight thermoplastic elastomeric block copolymer, 1 part by weight antioxidant, and 25 parts by weight tackifying resin. In Example 22, a pressure-sensitive adhesive  
 5 tape was prepared as in Example 20 except the proportions were 25 parts by weight acrylic adhesive, 37.5 parts by weight thermoplastic elastomeric block copolymer, 1 part by weight antioxidant, and 37.5 parts by weight tackifying resin. In Example 23, a pressure-sensitive adhesive was prepared as in Example 20 except the thermoplastic elastomeric block copolymer was KRATON™ D1112P  
 10 (styrene isoprene-styrene block copolymer) and the proportions were 50 parts by weight acrylic adhesive, 25 parts by weight thermoplastic elastomeric block copolymer, 1 part by weight antioxidant, and 25 parts by weight tackifying resin. Each adhesive was 25  $\mu\text{m}$  (1 mil) thick.

Comparative Example C10 was prepared as in Example 20, but contained  
 15 only the acrylic pressure-sensitive adhesive. The pressure-sensitive adhesive tapes were tested for 180° peel adhesion to glass, shear strength to fiberboard and polished steel and rolling ball tack. The results are set forth in Table 6.

Table 6

Example	Shear Strength (min)		Rolling Ball Tack ( $\mu\text{m}$ )	Peel Adhesion (N/dm)
	FB	SS		
C10	5	31	35	32
20	-	14	36	38
21	9	15	44	35
22	6	26	43	59
23	7	26	38	50

20

As can be seen from the data in Table 6, addition of the thermoplastic elastomeric block copolymer and tackifying resin improve the 180° peel adhesion of the acrylic pressure-sensitive adhesive with the greater improvement being seen in Example 22 of the adhesive compositions prepared with KRATON™ D1107P

as the thermoplastic elastomer block copolymer and a dramatic increase in peel  
adhesion being seen in the adhesive prepared with KRATON™ D1112 as the  
thermoplastic elastomeric block copolymer. The adhesives of Examples 20-23  
retained the excellent rolling ball tack of the adhesive containing only acrylic  
5 pressure-sensitive adhesive even when the amount of tackified thermoplastic  
elastomer was as high as 75 weight percent. In Example 20, the acrylic adhesive  
formed a continuous domain with the thermoplastic elastomer/tackifying resin  
forming a ribbon-like schistose structure. In Example 21 the structure was  
schistose in nature with the acrylic adhesive domain and the thermoplastic  
10 elastomer/tackifying resin domain being substantially co-continuous. In Example  
22, the thermoplastic elastomer/tackifying resin formed a substantially continuous  
domain with the acrylic adhesive being in a ribbon-like schistose form. FIG. 11  
shows the adhesive of Example 22 in cross-section in the film forming direction at  
2000X magnification after staining with osmium tetroxide. FIG. 12 shows the  
15 adhesive of Example 22 in cross-section along the cross-web direction after  
staining with osmium tetroxide. In Example 23 the structure was schistose in  
nature with the acrylic adhesive domain and the thermoplastic elastomer/tackifying  
resin domain being substantially co-continuous.

#### 20 Examples 24-27 and Comparative Examples C11 and C12

In Examples 24-27, an acrylic pressure-sensitive adhesive was prepared as  
described in Example 1. Core-sheath strands were prepared having a core of the  
acrylic adhesive and a sheath of thermoplastic elastomeric polymer (99 parts by  
weight KRATON™ D1107P) blended with an antioxidant (1 part by weight  
25 IRGANOX™ 1010, available from Ciba-Geigy Corp.). The weight ratios of  
acrylic adhesive, thermoplastic elastomer and antioxidant used in Examples 24-27  
was 70:30:1, 75:25:1, 80:20:1 and 70:30:1, respectively. The strands were  
quenched in water, dried and fed into a corotating twin screw extruder, Model  
ZSK 30, available from Werner & Pfleiderer Corp. In Examples 24-27, the  
30 adhesives were coated at coating weights of approximately 25 g/m<sup>2</sup> (6 gr/155 cm<sup>2</sup>)



onto a polyvinyl chloride-free film comprising a blend of 80 parts by weight ethylene-vinyl acetate and 20 parts by weight ethylene-propylene-diene rubber and a flame retardant system comprising 20 parts by weight alumina trihydrate, 11 parts by weight decabromodiphenyl oxide and 5 parts by weight antimony trioxide, prepared as described in U.S. Pat. No. 5,284,889 which is incorporated herein by reference. In Example 30, the adhesive was coated at a coating weight of 35 g/m<sup>2</sup> (8.4 gr/155 cm<sup>2</sup>) onto a polyvinyl chloride film backing designated 5A1Q801-1, available from Nanya Plastics Corp. In Comparative Example C11, the polyvinyl chloride-free tape backing was coated with a blend of 100 parts by weight thermoplastic elastomer (KRATON™ D1107P) and 1 part by weight antioxidant (IRGANOX™ 1010) at a coating weight of 75 g/m<sup>2</sup> (18 gr/155 cm<sup>2</sup>). In Comparative Example C12, the polyvinyl chloride-free backing was coated at a coating weight of 47 g/m<sup>2</sup> (11.4 g/155 cm<sup>2</sup>) using only the acrylic adhesive.

Peel adhesion of the adhesives to steel and backing at 20°C and -18°C was determined using the Electrical Tape Adhesion Test, the data from which was normalized to a coating weight of 29 g/m<sup>2</sup> (7 gr/155 cm<sup>2</sup>) using the formula

$$\text{Normalized Adhesion} = \frac{\text{Measured Adhesion} \times 0.45}{\text{Measured Coating Weight (g)}}$$

where coating weight is the weight of adhesive per 155 cm<sup>2</sup> (4 in x 6 in) area. The results are set forth in Table 7.

Table 7

Example Adhesion (N/dm)	C11	C12	24	25	26	27
Steel @ 20°C	0.7	12	17	26	23	20
Backing @ 20°C	0.4	5.2	4.7	7.5	6.5	22
Steel @ -18°C	0	26	38	54	45	-
Backing @ -18°C	0	13	31	41	37	-

As can be seen from the data in Table 7, the pressure-sensitive adhesive tapes of the present invention, Examples 24-27, perform better than either component alone on steel at 20°C and generally on backing at 20°C. At -18°C, the pressure-sensitive electrical tapes of Examples 24-27 performed significantly better than the tapes of Comparative Examples C11 and C12.

During coating of the adhesive materials in Examples 24-26, approximately 1 to 2 mm thick samples of adhesive were collected from the die onto a polyester release liner. These samples were examined by dynamic mechanical analysis using a RHEOMETRIC™ dynamic analyzer (available from Rheometric Scientific, Inc.). FIG. 14 shows the dynamic mechanical properties  $G'(\omega)$  and  $\tan^2\delta$  obtained from this measurement, where dashed lines represent Example 24, dotted lines represent Example 25 and solid lines represent Example 26. The compliance values were calculated using the equation  $J(t) = 1/(G'(\omega) \times (1 + \tan^2\delta))$  where  $G'(\omega)$  is the shear storage modulus equal to (tensile storage modulus)/3 and  $J(t)$  is the shear creep compliance, a measure of the softness of the adhesive. The higher the  $J(t)$  value, the better the contact between the adhesive and the surface to which it is bonded. The results in  $\text{cm}^2/\text{dyne}$  are set forth in Table 8.

Table 8

Example	24	25	26
Tan $\delta$ @ -20°C	0.79	0.82	1.13
@ -10°C	0.39	0.44	0.56
@ 23°C	0.30	0.33	0.32
$G'(\text{dynes}/\text{cm}^2)$ @ -20°C	$5.0 \times 10^6$	$4.3 \times 10^6$	$6.5 \times 10^6$
@ -10°C	$3.0 \times 10^6$	$2.0 \times 10^6$	$2.9 \times 10^6$
@ 23°C	$7.8 \times 10^5$	$6.4 \times 10^5$	$7.5 \times 10^5$
$J(t) (\text{cm}^2/\text{dyne})$ @ -20°C	$1.6 \times 10^{-7}$	$1.8 \times 10^{-7}$	$1.0 \times 10^{-7}$
@ -10°C	$3.1 \times 10^{-7}$	$4.6 \times 10^{-7}$	$3.0 \times 10^{-7}$
@ 23°C	$12.0 \times 10^{-7}$	$15.0 \times 10^{-7}$	$13.0 \times 10^{-7}$

As can be seen from the data in Table 8, each of the pressure-sensitive adhesive tapes of Examples 24-26 has excellent compliance, with the best performance shown by the tape of Example 25. Such compliance, or softness, indicates that the adhesive of this invention would make good contact with a surface to which it is bonded and that the adhesive would generally have good peel adhesion.

#### Example 28 and Comparative Example C13

Pressure-sensitive adhesive tapes were prepared as in Example 15 and Comparative Example C9. Samples of each tape were subjected to ultraviolet irradiation using a UV Processor Model #QC120244ANIR available from RPC Industries, Inc. with medium mercury lamps. The irradiation was performed in an inert nitrogen atmosphere at a calibrated dose of 100 mJ/cm<sup>2</sup> at 365 nm. The tapes were tested for 180° peel adhesion and for shear strength. The results are set forth in Table 9 together with the results of the tests on Example 15 and Comparative Example C9.

Table 9

Example	Peel Adhesion (N/dm)	Shear Strength (min) (Failure Mode)
C9	31.5	27 (cohesive)
15	55.3	37 (cohesive)
C13	37.1	21 (adhesive: pop-off)
28	71.7	216 (adhesive: pop-off)

Radiation curing with ultraviolet light to increase crosslinking, caused greater increases in peel adhesion with the tape prepared with the adhesive of the invention. However, the most significant increase due to irradiating the samples was seen in the shear strength where the irradiated adhesive of the invention increased in shear strength more than five times over the non-irradiated tape, while the irradiated tape prepared with only acrylate adhesive decreased in shear strength after irradiation.

Examples 29-35 and Comparative Examples C14-C15.

The acrylic pressure-sensitive adhesive described in Example 1 was blended with KRATON™ D1107P at various ratios and coated using batch mixing and coating processes. For Examples 29-35, a total of 45 grams of acrylic pressure-sensitive adhesive and KRATON™ D1107P (99 parts preblended with 1 part IRGANOX™ 1010 antioxidant) were charged into a 50 gram Brabender™ sigma blade mixer operating at 175°C (347°F). The KRATON™ D1107P was added first, followed by the acrylic pressure-sensitive adhesive. The blend was mixed at 50-75 rpm for 5 minutes. The relative amounts of acrylic pressure-sensitive adhesive and KRATON™ D1107P that were blended correspond to 40.5 grams and 4.5 grams for Example 29, 33.75 grams and 11.25 grams for Example 30, 22.5 grams and 22.5 grams for Example 31, 18 grams and 27 grams for Example 32, 11.25 grams and 33.75 grams for Example 33, 6.75 grams and 38.25 grams for Example 34, and 2.25 grams and 42.75 grams for Example 35.

Pressure-sensitive adhesive coated tapes were prepared for each example by placing approximately 1-5 grams of blended material between a sheet of release coated paper and a 50-75 micron thick biaxially oriented polyethylene terephthalate film. This was then placed between two aluminum plates in a hydraulic platen press having two steel plates measuring 152 mm (6 inches) by 152 mm (6 inches). Application of 28 MPa (4000 psi) at 138°C (280°F) and 0.6 second dwell time produced pressure-sensitive adhesive coatings having thickness ranging from 76 µm (3 mils) to 102 µm (4 mils). The tapes were tested for 180° peel adhesion to glass. The results are set forth in Table 10.

Table 10

Example	Acrylic Pressure-sensitive Adhesive (Wt. %)	KRATON™ D1107P (Wt. %)	180° Peel (N/dm)
C14	100	0	61
29	90	10	71
30	75	25	81
31	50	50	81
32	40	60	59
33	25	75	49
34	15	85	29
35	5	95	26
C15	0	100	0

The morphologies of the adhesives of Examples 29 and 30 were a continuous acrylic pressure-sensitive adhesive phase with schistose thermoplastic elastomer phase. The adhesives of Examples 31 and 32 had co-continuous schistose phases of acrylic pressure-sensitive adhesive and thermoplastic elastomer. Examples 33-35 had a continuous non-tacky thermoplastic elastomer phase with discontinuous pressure-sensitive adhesive schistose phase. Due to the axial deformation of the platen press coating method, the phases are formed parallel to the plane of the coating. As can be seen from Table 10, the 180° peel adhesion to glass can be controlled by varying the ratio of acrylic pressure-sensitive adhesive to thermoplastic elastomer. Behavior ranging from enhanced 180° peel adhesion (Examples 29-31) to reduced 180° peel adhesion and easier removal (Examples 32-35) is demonstrated.

#### Examples 36 - 37

Acrylic pressure-sensitive adhesive tapes were prepared as in Example 29 except different acrylic pressure-sensitive adhesives were used and the ratio of



acrylic pressure-sensitive adhesive to thermoplastic elastomer was 80:20. For Example 36, the acrylic pressure-sensitive adhesive used was 2-ethylhexyl acrylic pressure-sensitive adhesive prepared by feeding a premix of isooctylacrylate : acrylic acid : isooctylthioglycolate : VAZO™ 64 (available from DuPont) in a ratio of 90:10:0.05:0.3 into an 18 mm Leistritz counterrotating twin screw extruder equipped with fully intermeshing, integrated screws according to the general method described in U.S. Pat. No. 4,619,979 (Kotnour, et al) which is incorporated herein by reference. The extruder was not vented and the screw speed was maintained at 50 rpm. The temperatures in zones 1 through 8 were maintained at 72°C, 82°C, 92°C, 96°C, 100°C, 110°C, 115°C and 120°C, respectively. For Example 37, the acrylic pressure-sensitive adhesive used was n-butyl acrylic pressure-sensitive adhesive prepared in a similar manner as the 2-ethyl hexyl acrylic pressure-sensitive adhesive of Example 36 except the materials used were n-butyl acrylate : N,N-dimethylacrylamide : acrylic acid : 4-acryloxy-benzophenone : isopropanol : and VAZO™ 64 in a ratio of 83:15:2:0.2:2:0.3, the extruder rpm was 70 and temperatures in zones 1 through 8 were maintained at 70°C, 81°C, 92°C, 96°C, 105°C, 110°C, 115°C and 120°C, respectively. For each example, the relative amount of acrylic pressure-sensitive adhesive and preblended KRATON™ D1107P and IRGANOX™ 1010 blended at 100 parts thermoplastic elastomer to 2 parts antioxidant. Comparative Examples C16 and C17 were made as in Examples 36 and 37, respectively, except only the acrylic pressure-sensitive adhesive was used with no thermoplastic elastomer. The samples were tested for 180° peel adhesion from glass and the adhesive thickness and peel adhesion values are listed in Table 10.

25

Table 10

Example	Adhesive Thickness ( $\mu\text{m}$ )	Peel Adhesion (N/dm)
36	100	78
C16	100	63
37	100	108
C17	100	92

As can be seen by the data in Table 10, the adhesive compositions prepared from the acrylic pressure-sensitive adhesives and the thermoplastic elastomer block copolymer had superior 180° peel adhesion values than the comparative examples containing no thermoplastic elastomer. The morphologies of the adhesive compositions were a continuous acrylic pressure-sensitive adhesive phase with a schistose thermoplastic elastomer phase.

#### 10 Example 38

In Example 38, pressure-sensitive adhesive tape was prepared as in Example 1 with the acrylic pressure-sensitive adhesive used in Example 1 and with the thermoplastic elastomeric polymer HYTREL™ 5326, a polyetherester available from DuPont Co., shear viscosity - 495 Pa-s, which had been preblended with 1 weight percent IRGANOX™ 1010, such that the ratio of acrylic adhesive to thermoplastic elastomeric polymer was 80:20. The adhesive thickness was 60  $\mu\text{m}$  and the 180° peel adhesion value from glass was 40 N/dm. The morphology of the adhesive was a continuous acrylic pressure-sensitive adhesive phase with a schistose thermoplastic elastomer phase.

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#### Examples 39-42 and Comparative Examples C18 and C19

In Examples 39, 40 and 41, the water suspension polymerized acrylic pressure-sensitive adhesive described in Example 12 and 13 was melt blended with a thermoplastic elastomeric adhesive (prepared by blending 50 parts thermoplastic elastomeric block copolymer KRATON™ D1107P, 1 part antioxidant

25

IRGANOX™ 1010 and 50 parts tackifying resin ESCOREZ™ 1310LC) in a corotating twin screw extruder, Model ZSK 30, having 30 mm diameter barrel and a length to diameter ratio of 37:1 with the acrylic adhesive to thermoplastic elastomer adhesive ratio being 75:25, 50:50 and 25:75, respectively. The thermoplastic elastomer block copolymer was fed into zone 1, the tackifying resin in zone 2 and the acrylic pressure-sensitive adhesive in zone 3. Temperatures were maintained between 149°C and 165°C. In Example 42, a pressure-sensitive adhesive tape was prepared as in Example 39 using the water emulsion polymerized acrylic pressure-sensitive adhesive described in Example 1 and the thermoplastic elastomer described in Example 29 with the acrylic pressure-sensitive adhesive to thermoplastic elastomer block copolymer ratio being 75:25. In Comparative Example C18, the pressure-sensitive adhesive tape was prepared using only acrylic adhesive. In Comparative Example C19, the pressure-sensitive adhesive tape was prepared using only the tackified thermoplastic elastomeric adhesive. All samples had an adhesive coating thickness of about 50 µm (2 mils) and were coated onto non-occlusive, i.e. breathable, woven backing which has an 180 x 48 plain weave acetate taffeta cloth, 75 denier fiber in the warp direction and 150 denier fiber in the fill direction, available from Milliken and Co., Spartanburg, GA. The adhesive compositions in Example 39 and 42 showed a substantially continuous acrylic adhesive domain with the thermoplastic elastomer/tackifying resin forming schistose ribbon-like domains. In the adhesive composition of Example 40, the acrylic adhesive and the thermoplastic elastomer/tackifying resin formed substantially co-continuous schistose domains. In Example 41, the thermoplastic elastomer/tackifying resin formed a substantially continuous domain, while the acrylic adhesive formed schistose ribbon-like domains. FIG. 15 shows the adhesive of Example 40 in cross-section in the film forming direction after staining with osmium tetroxide. The pressure-sensitive adhesive tapes were tested for skin adhesion immediately after application,  $T_0$ , and after 24 hours,  $T_{24}$ , skin adhesion lift after 24 hours and skin adhesion residue after 24 hours. The results are set forth in Table 11.

Table 11

Example	T <sub>0</sub> (N/dm)	T <sub>24</sub> (N/dm)	T <sub>24</sub> Lift	T <sub>24</sub> Residue
C18	2.2	7.9	1.8	1.0
39	3.0	11.2	1.6	0.8
40	4.0	7.7	1.6	0.6
41	3.4	4.3	1.6	0.3
C19	3.0	3.3	1.3	0.1
42	2.0	2.7	0.3	1.0

As can be seen from the data in Table 11, the pressure-sensitive adhesive tapes on Examples 39-42 had enhanced peel performance from skin and the T<sub>0</sub>:T<sub>24</sub> adhesion can be controlled by appropriate blending of the acrylic adhesive and the tackified or untackified thermoplastic elastomer. In particular, the tape of Example 40 had between 180 percent and 33 percent higher initial adhesion to skin than tapes prepared of either of the component pressure-sensitive adhesives, Comparative Examples C18 and C19. Additionally, all Examples provided adhesives with acceptable 24 hour aged adhesion to skin.

#### Examples 43-45

In Example 43, the acrylic pressure-sensitive adhesive used in Example 12 was melt-blended with a thermoplastic elastomeric adhesive (prepared by preblending 100 parts thermoplastic elastomeric block copolymer KRATON™ D1107P, 1.5 parts antioxidant IRGANOX™ 1076, available from Ciba-Geigy Corp, 1.5 parts antioxidant CYANOX™ LTDP, available from American Cyanamide Corp., Wayne, NJ, and 70 parts tackifying resin WINGTACK™ Plus, available from Goodyear Chemical Co., Akron, OH) with the acrylic adhesive to thermoplastic elastomer adhesive ratio being 65:35 using an 8.9 cm diameter screw pin barrel mixer, available from The French Oil Mill Machinery Co., Piqua, OH, with zone temperatures rising from 106°C to 144°C and water injected at 1

part per 100 parts pressure-sensitive adhesive composition as the composition leaves the pin barrel mixer. A gear pump attached to the output end of the pin barrel mixer by a heated pipe delivered the pressure-sensitive adhesive composition to a wipe-film coating die, maintained at a temperature of 160°C, for coating the adhesive composition at a rate of 1.8 kg/hour/25 cm die width onto a non-occlusive woven backing as used in Examples 39-42. The backing speed was adjusted to provide adhesive coatings having an average thickness of about 50 µm (2 mils). In Example 44, a pressure-sensitive adhesive tape was prepared as in Example 42 except the thermoplastic elastomeric adhesive was prepared by blending 50 parts thermoplastic elastomeric block copolymer KRATON™ D1119, a styrene-isoprene-styrene block copolymer, shear viscosity - 17 Pa-s, available from Shell Chemical Co), 2 parts antioxidant IRGANOX™ 1076 and 48 parts tackifying resin WINGTACK™ Plus. In Example 45, a pressure-sensitive adhesive tape was prepared as in Example 43 except the acrylic pressure-sensitive adhesive used in Example 1 was melt-blended with a thermoplastic elastomeric adhesive, prepared by blending 50 parts thermoplastic elastomeric block copolymer KRATON™ D1107P, 1 part antioxidant IRGANOX™ 1010, and 50 parts tackifying resin ESCOREZ™ 1310LC, with the acrylic adhesive to thermoplastic elastomer adhesive ratio being 25:75, the pressure-sensitive adhesive composition being fed at 15.1 Kg/hr and the zone temperatures maintained between 124°C and 131°C. The adhesive compositions in Example 43 and 44 showed a substantially continuous acrylic adhesive domains with the thermoplastic elastomer/tackifying resin forming schistose ribbon-like domains. The adhesive compositions in Example 45 showed a substantially continuous thermoplastic elastomer/tackifying resin domain with the acrylic adhesive forming schistose ribbon-like domains. The pressure-sensitive adhesive tapes were tested for skin adhesion immediately after application,  $T_0$ , and after 48 hours,  $T_{48}$ , skin adhesion lift after 48 hours and skin adhesion residue after 48 hours. The results are set forth in Table 12.



Table 12

Example	T <sub>0</sub> (N/dm)	T <sub>48</sub> (N/dm)	T <sub>48</sub> Lift	T <sub>48</sub> Residue
43	4.9	12.0	1.9	1.6
44	4.6	10.8	1.9	1.7
45	2.1	5.4	0.7	0.3

As can be seen from the data in Table 12, the pressure-sensitive adhesive tapes on Examples 43, 44 and 45, with non-occlusive woven backings and with  
5 different acrylic pressure-sensitive adhesives and thermoplastic elastomeric adhesives, had acceptable peel performance from skin.

#### Examples 46-48

In Examples 46-48, pressure-sensitive adhesive tapes were made with  
10 various non-occlusive backings using the same thermoplastic elastomer/tackifying resin adhesive, melt-mixing and coating process as in Example 45. The acrylic adhesive was as used in Example 12 and 13. In Example 46, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 60:40 and the pressure-sensitive adhesive composition was coated onto a release liner and laminated to a  
15 nonwoven rayon fiber backing. The backing was formed by first passing 2.5 to 5 cm long, 1.5 denier viscose-rayon staple fibers through a twin cylinder card (available from Spinnbau GmbH, Bremen, Germany) to form a fluffy fiber web with a fiber weight of between 41 g/m<sup>2</sup> and 54 g/m<sup>2</sup>. The fluffy fiber web was simultaneously compacted to a tissue-like condition and sized by being fed  
20 through the nip of a pair of horizontal squeeze rolls, the lower one of which dips in an aqueous bath of fiber-binding rubbery acrylate sizing latex (RHOPLEX™ B-15, available from Rohm and Haas Co.), diluted with water to provide a size weight approximately equal to the weight of the fiber; and then dried. In Example 47, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 50:50 and  
25 the pressure-sensitive adhesive composition was applied to a liner and melt blown microfiber was blown onto the adhesive at 450 g/hr/cm to form an 80 μm thick

backing with a basis weight of 20 g/m<sup>2</sup>. The melt blown microfibers had a diameter of between about 5 and 10  $\mu$ m and were prepared using PS 440-200 polyurethane, available from Morton International, Inc., Seabrook, NH, and a process similar to that describe in U. S. Pat. No. 5,230,701, Example 1. In

5 Example 48, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 60:40 and the pressure-sensitive adhesive composition was applied to 0.65 mm thick SONTARA™ 8010 backing, a 44 g/m<sup>2</sup> basis weight hydroentangled polyester nonwoven substrate available from DuPont Co. In the adhesive composition of Example 46, 47 and 48, the acrylic adhesive and the thermoplastic

10 elastomer/tackifying resin adhesive formed substantially co-continuous schistose domains. The pressure-sensitive adhesive tapes were tested for skin adhesion immediately after application, T<sub>0</sub>, and after 48 hours, T<sub>48</sub>, skin adhesion lift after 48 hours and skin adhesion residue after 48 hours. The thickness of each of the adhesive composition examples and the test results are set forth in Table 13.

15

Table 13

Example	Thickness ( $\mu$ m)	T <sub>0</sub> (N/dm)	T <sub>48</sub> (N/dm)	T <sub>48</sub> Lift	T <sub>48</sub> Residue
46	21	2.1	6.5	0.1	0.6
47	39	2.6	6.1	0.5	0.0
48	32	3.5	12.0	0.9	4.8

As can be seen from the data in Table 13, the pressure-sensitive adhesive tapes on Examples 46, 47 and 48, with non-occlusive woven backings and with

20 different acrylic pressure-sensitive adhesives and thermoplastic elastomeric adhesives, had acceptable peel performance from skin.

#### Examples 49-52

In Examples 49-52, pressure-sensitive adhesive tapes were made as in

25 Examples 46-48 except various occlusive non-breathable backings were used. In Example 49, the acrylic adhesive to thermoplastic elastomer adhesive ratio was

60:40 and the pressure-sensitive adhesive composition was applied to a 117  $\mu\text{m}$  thick polyethylene/vinyl acetate copolymer film prepared using ESCORENE™ :S-31209, available from Exxon Chemical Co. The film was perforated with about 100 holes/cm<sup>2</sup>. In Example 50, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 60:40 and the pressure-sensitive adhesive composition was applied to a 76  $\mu\text{m}$  thick low density polyethylene film, prepared using NA 964-085 resin, available from Quantum Chemical Co. In Example 51, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 50:50 and the pressure-sensitive adhesive composition was applied to 0.57 mm thick plasticized polyvinyl chloride foam (available as No. 9058 TA 022 Fleshtone from General Foam Corp., Carlstat, NJ. In Example 52, the acrylic adhesive to thermoplastic elastomer adhesive ratio was 50:50 and the pressure-sensitive adhesive composition was applied to the polymer side of a white polymer/cloth laminate composed of ENGAGE™ 8200 (a polyolefin elastomer available from Dow Plastics Co) extrusion coated onto 44x36 woven cloth (available from Burcott Mills). White backing was produced by dry blending 1 part of 50:50 titanium dioxide in low density polyethylene (available as PWC00001 from Reed Spectrum, Holden MA) with 3 parts ENGAGE™ 8200; forming pigmented pellets by melt mixing the blend in a 40 mm twin screw extruder (available from Berstorff) at 200°C and extruding and pelletizing strands; dry blending the pigmented pellets with more unpigmented ENGAGE™ 8200 in a ratio of 1:25; melt mixing the blend and feeding the blend at approximately 270 g/min into the feed throat of a 6.4 cm diameter Davis Standard Model N 9485 Single Screw Extruder, available from Davis Standard Div., Crompton and Knowles Corp., Pawcatuck, CT, at 204°C (400°F) and extruding a 66  $\mu\text{m}$  (2.6 mil) thick film onto the cloth with the casting roll temperature set at 93°C (200°F) to form a laminate; and passing the laminate through the nip of two horizontal rolls at a pressure of 350 N/cm at approximately 11 m/min (approximately 35 fpm). In the adhesive composition of Examples 49, 50 and 52, the acrylic adhesive and the thermoplastic elastomer/tackifying resin adhesive formed substantially co-continuous schistose

domains. In the adhesive composition of Example 51, the acrylic adhesive formed a substantially continuous domain and the thermoplastic elastomer/tackifying resin adhesive formed ribbon-like schistose domains. The pressure-sensitive adhesive tapes were tested for skin adhesion immediately after application,  $T_0$ , and after 48 hours,  $T_{48}$ , skin adhesion lift after 48 hours and skin adhesion residue after 48 hours. The thickness of each of the adhesive composition examples and the test results are set forth in Table 14.

Table 14

Example	Thickness ( $\mu\text{m}$ )	$T_0$ (N/dm)	$T_{48}$ (N/dm)	$T_{48}$ Lift	$T_{48}$ Residue
49	29	2.4	1.8	1.3	0.6
50	39	2.1	1.5	0.9	0.6
51	39	5.6	7.6	0.3	1.8
52	50	1.5	3.7	1.1	0.3

As can be seen from the data in Table 14, the pressure-sensitive adhesive tapes of Examples 49 to 52 with occlusive nonbreathable backings had varying peel performance from skin.

#### 15 Example 53

Pressure-sensitive adhesive tape was prepared using the procedure of Example 18 and the materials described in Example 52 except 10 parts MICRAL™ 1500, an alumina trihydrate, available from Salem Industries, Inc., Norcross, GA, per 100 parts thermoplastic elastomeric adhesive was preblended with the thermoplastic elastomeric adhesive. The pressure-sensitive adhesive had a thickness of about 50  $\mu\text{m}$ . The acrylic adhesive and the thermoplastic elastomer/tackifying resin adhesive formed substantially co-continuous schistose domains. The pressure-sensitive adhesive tape was tested for skin adhesion immediately after application,  $T_0$ , and after 48 hours,  $T_{48}$ , skin adhesion lift after 48 hours and skin adhesion residue after 48 hours. The skin adhesion was 1 N/dm

initially and 3.1 N/dm after 48 hours. After 48 hours the skin adhesion lift was 1 and the skin adhesion residue was 0.

The various modifications and alterations of this invention will be apparent  
5 to those skilled in the art without departing from the scope and spirit of this invention and this invention should not be restricted to that set forth herein for illustrative purposes only.



What is claimed is:

1. A pressure-sensitive adhesive composition comprising a blend of about 5 to 95 weight percent of an acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of a thermoplastic elastomeric copolymer, said composition having  
5 a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to a major surface of the adhesive composition within said first domain.
- 10 2. The pressure-sensitive adhesive composition of claim 1 wherein the acrylic pressure-sensitive adhesive comprises a polymer of a C<sub>3</sub>-C<sub>12</sub> alkyl ester.
3. The pressure-sensitive adhesive composition of claim 2 wherein the acrylic pressure-sensitive adhesive comprises a polymer of isooctyl acrylate, 2-  
15 ethyl-hexyl acrylate or n-butyl acrylate.
4. The pressure-sensitive adhesive composition of claim 2 wherein the acrylic pressure-sensitive adhesive further comprises a polar component.
- 20 5. The pressure-sensitive adhesive composition of claim 4 wherein the polar component comprises acrylic acid, methacrylic acid, ethylene vinyl acetate, N-vinyl pyrrolidone and styrene macromer.
6. The pressure-sensitive adhesive composition of claim 5 wherein the  
25 acrylic pressure-sensitive adhesive comprises about 100 to 80 weight percent alkyl ester component and about 0 to 20 weight percent polar component.
7. The pressure-sensitive adhesive composition of claim 1 wherein the thermoplastic elastomeric materials comprise linear, radial, star, tapered or  
30 branched copolymers.

8. The pressure-sensitive adhesive composition of claim 7 wherein the thermoplastic elastomeric materials comprise styrene-isoprene block copolymers, styrene-(ethylene-butylene) block copolymers, styrene-(ethylene-propylene) block copolymers, styrene-butadiene block copolymers, polyetheresters and poly- $\alpha$ -olefins.

9. The pressure-sensitive adhesive composition of claim 1 further comprising a tackifying resin.

10. The pressure-sensitive adhesive composition of claim 9 wherein said tackifying resin comprises up to 200 weight percent based on the weight of the thermoplastic elastomeric material.

11. A process for preparing a pressure-sensitive adhesive comprising the steps of

- (1) melt blending 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and 5 to 95 weight percent of at least one thermoplastic elastomeric material,
- (2) (a) forming said melt blended materials under shear or extensional conditions or both or  
(b) forming and drawing said melt blend  
to form a pressure-sensitive adhesive composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to the major surface of the adhesive within said first domain, and
- (3) allowing said composition to cool.

12. The process of claim 11 wherein the melt blending is carried out under dispersive or distributive conditions or a combination thereof.

13. The process of claim 11 wherein the mixing is carried using either a batch or continuous process.

14. The process of claim 13 wherein the batch process is carried out using  
5 internal mixing or roll milling.

15. The process of claim 13 wherein the continuous process is carried out using a single screw extruder, a twin screw extruder, a disk extruder, a reciprocating single screw extruder or a pin barrel single screw extruder.

10

16. A pressure-sensitive adhesive electrical tape comprising a polyvinyl chloride substrate or a substrate film of a blend of ethylene-vinyl acetate and ethylene-propylene-diene rubber and on the substrate, a pressure-sensitive adhesive composition comprising a blend of about 5 to 95 weight percent of an  
15 acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of a thermoplastic elastomeric block copolymer, said composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to a major surface of the adhesive composition within  
20 said first domain.

17. A pressure-sensitive adhesive tape comprising a substrate and on the substrate the pressure-sensitive adhesive composition of claim 1.

25 18. A double coated pressure-sensitive adhesive tape comprising a substrate and on at least one face of the substrate the pressure-sensitive adhesive composition of claim 1 and on the other surface of said substrate, a pressure-sensitive adhesive.

19. A process for preparing a pressure-sensitive adhesive tape comprising the steps of

- 5 (1) melt blending 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and 5 to 95 weight percent of at least one thermoplastic elastomeric material,
- (2) (a) forming said melt blended materials under shear or extensional conditions or both or
- (b) forming and drawing said melt blend,
- 10 said blend forming an adhesive composition and said composition being extruded onto a substrate to form a pressure-sensitive adhesive tape, said adhesive having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and
- 15 (3) allowing said adhesive to cool.

20. A process for preparing a pressure-sensitive adhesive tape comprising the steps of

- 20 (1) melt blending 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and 5 to 95 weight percent of at least one thermoplastic elastomeric material,
- (2) (a) forming said melt blended materials under shear or extensional conditions or both or
- (b) forming and drawing said melt blend
- 25 to form a pressure-sensitive adhesive composition and coextruding a film forming polymeric resin with said adhesive composition, said adhesive composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and
- 30

- (3) allowing said composition and said polymeric resin to cool.

21. A process for forming a double coated pressure-sensitive adhesive tape comprising the steps of

- 5 (1) melt blending 5 to 95 weight percent of at least one acrylic pressure-sensitive adhesive and 5 to 95 weight percent of at least one thermoplastic elastomeric material,
- (2) (a) forming said melt blended materials under shear or extensional conditions or both or
- 10 (b) forming and drawing said melt blend,
- to form a pressure-sensitive adhesive composition, said composition being fed to the outer portions of a feed block having at least three layers and a film forming polymeric resin being fed to a middle portion of said feed block to form a double-coated pressure-sensitive tape, said adhesive
- 15 composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature in the adhesive forming direction within said first domain, and
- (3) allowing said tape to cool.

20

22. A pressure-sensitive adhesive tape for adhesion to skin comprising a substrate and on the substrate a pressure-sensitive adhesive composition comprising a blend of about 5 to 95 weight percent of acrylic pressure-sensitive adhesive and about 5 to 95 weight percent of thermoplastic elastomeric material,
- 25 said composition having a morphology comprising at least two distinct domains, a first domain being substantially continuous in nature and said second domain being fibrillose to schistose in nature parallel to the major surface of the adhesive within said first domain.



23. The tape of claim 22 wherein said substrate is occlusive or non-occlusive.

24. The tape of claim 23 wherein said occlusive backing is a film, a  
5 foam material or laminate thereof.

25 The tape of claim 23 wherein said non-occlusive backing is a  
perforated polymeric film, a foam material, a woven fabric or a nonwoven fabrics.

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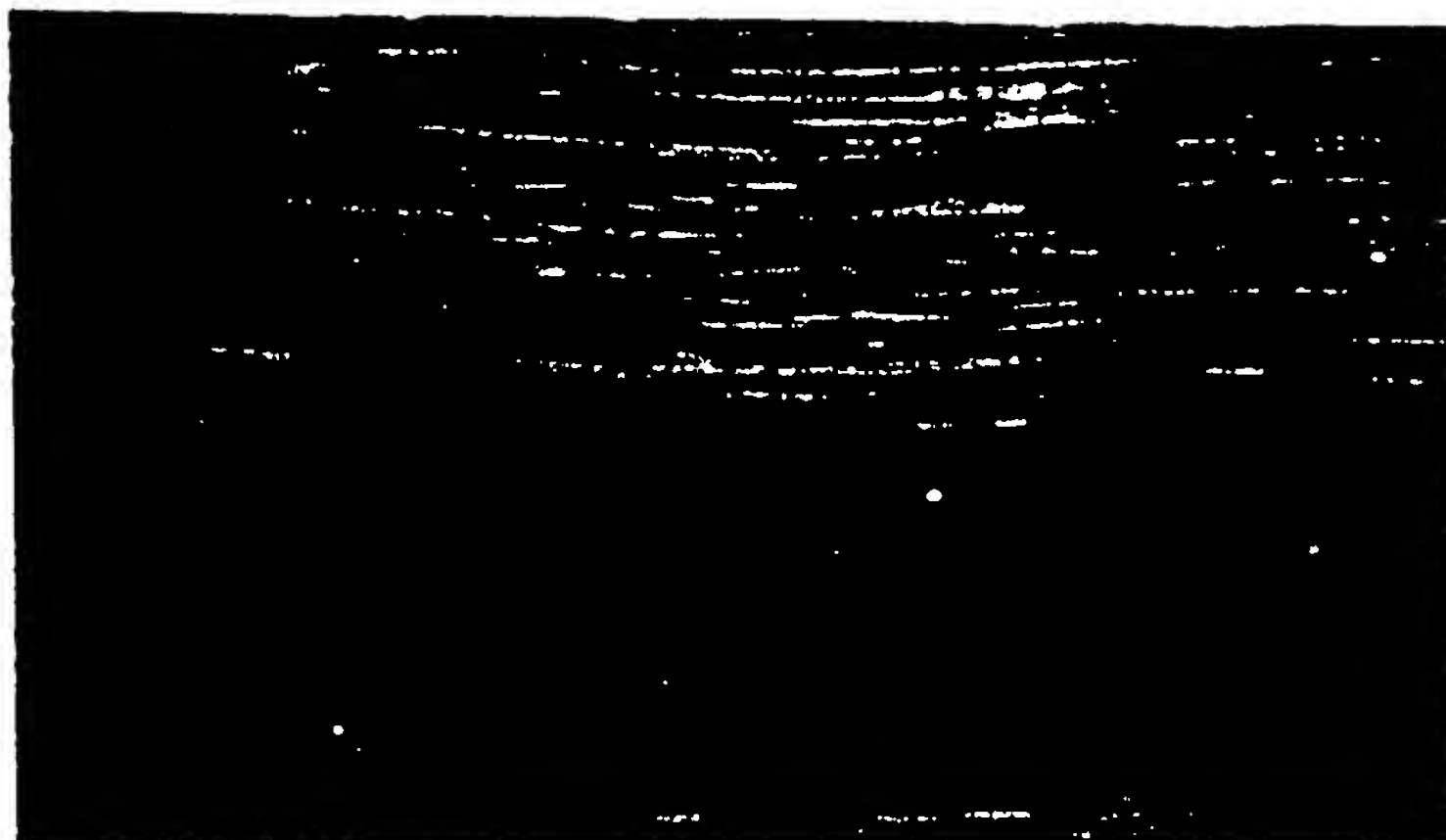


FIG. 1

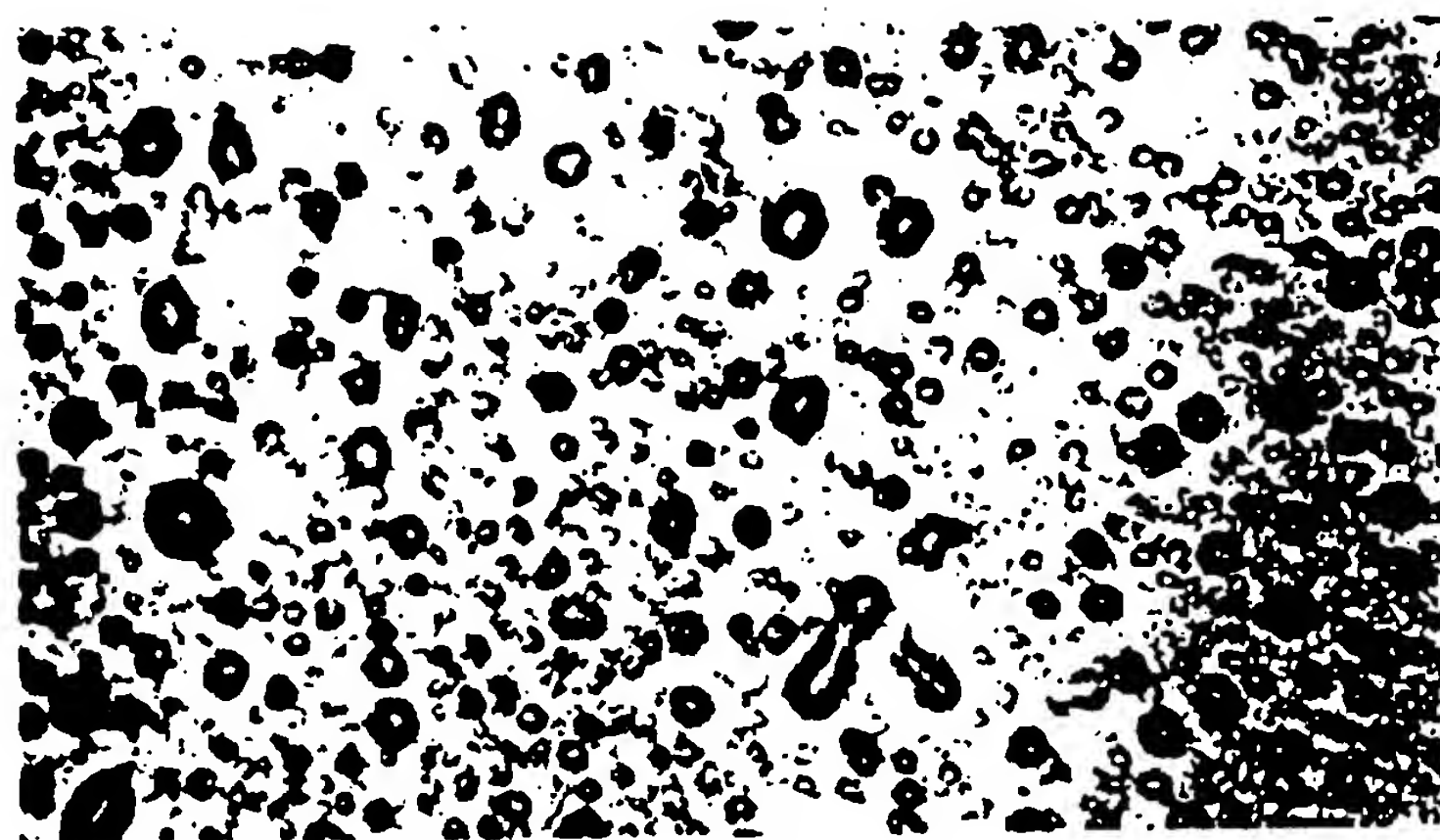


FIG. 2

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FIG. 3

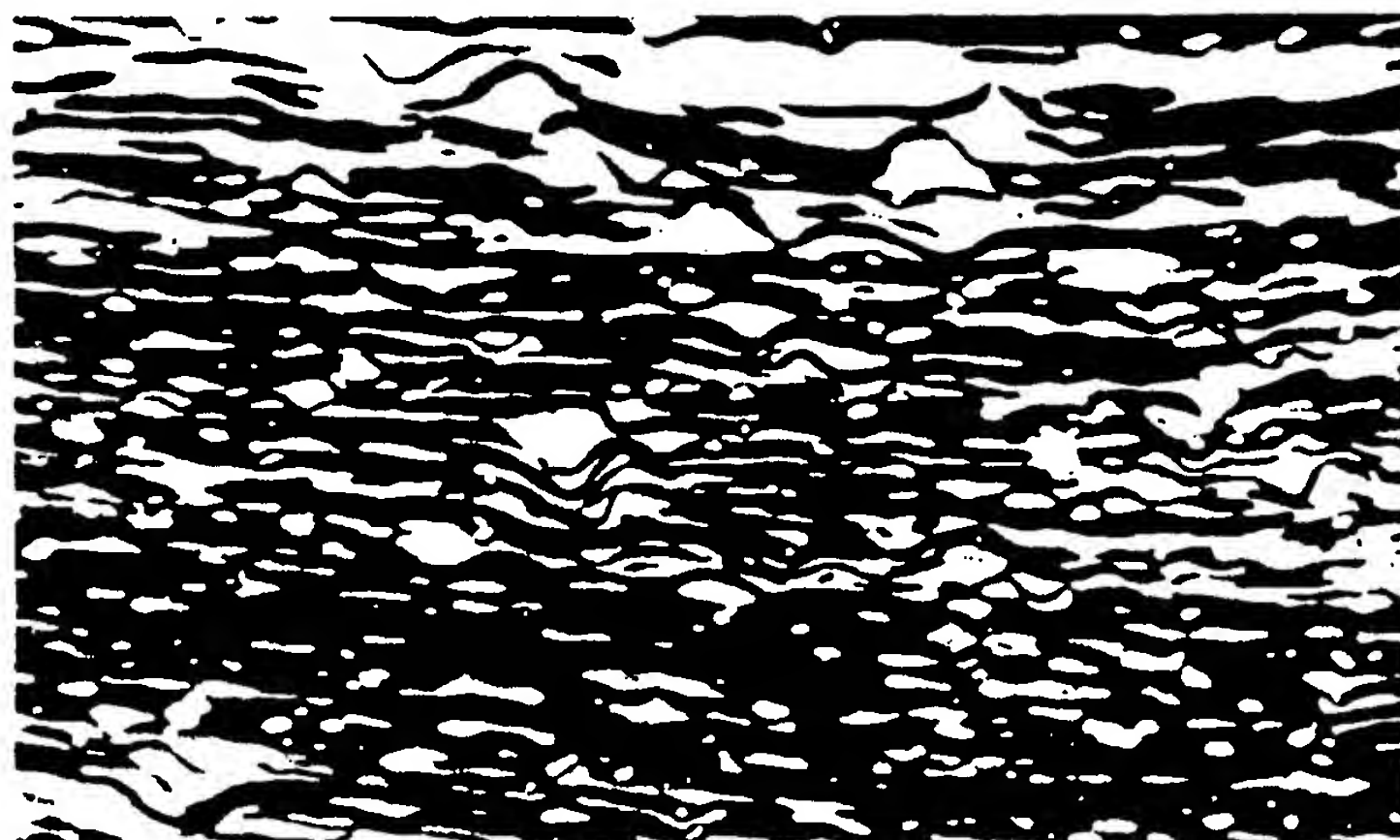


FIG. 4

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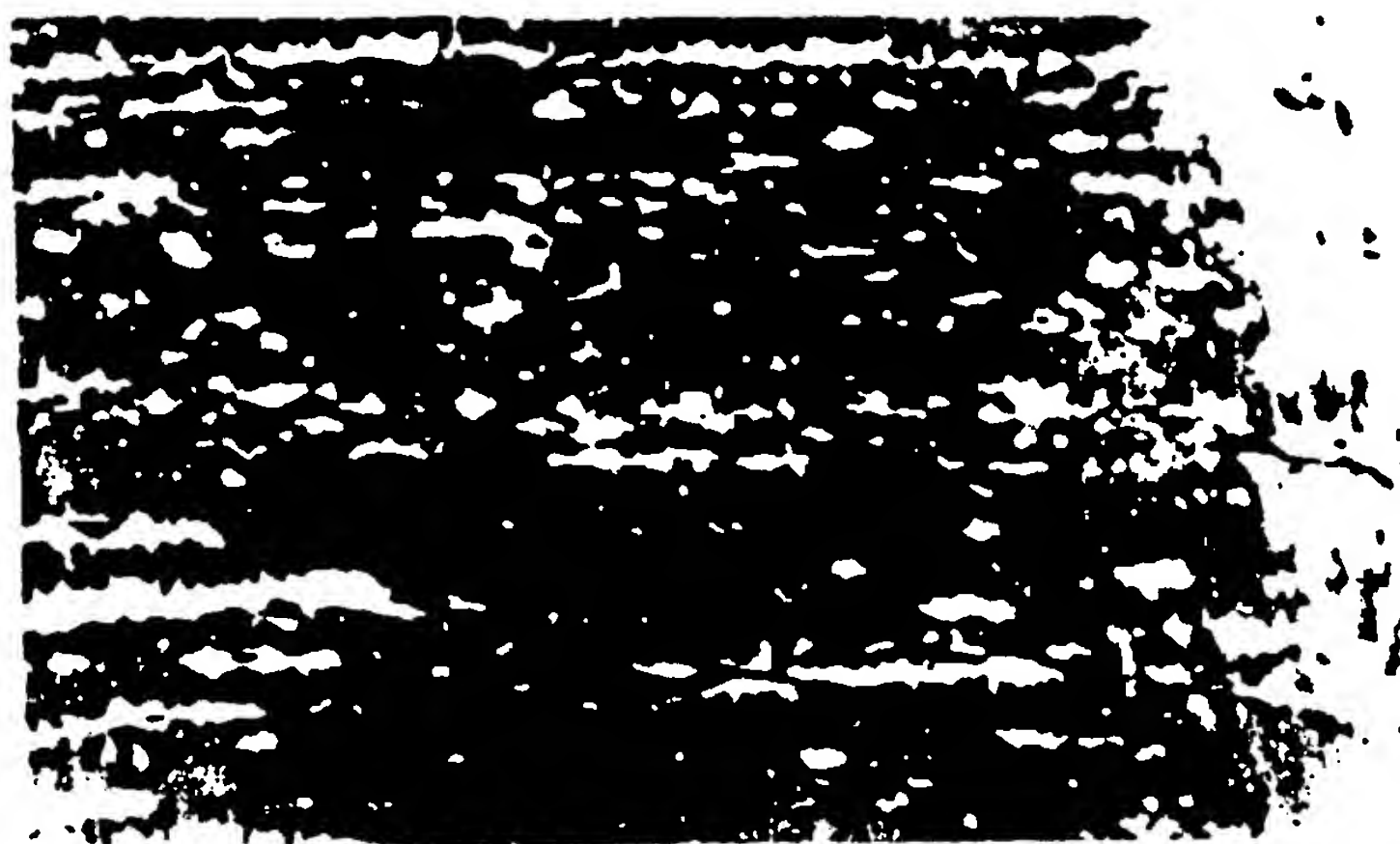


FIG. 5



FIG. 6

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FIG. 7



FIG. 8



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FIG. 9

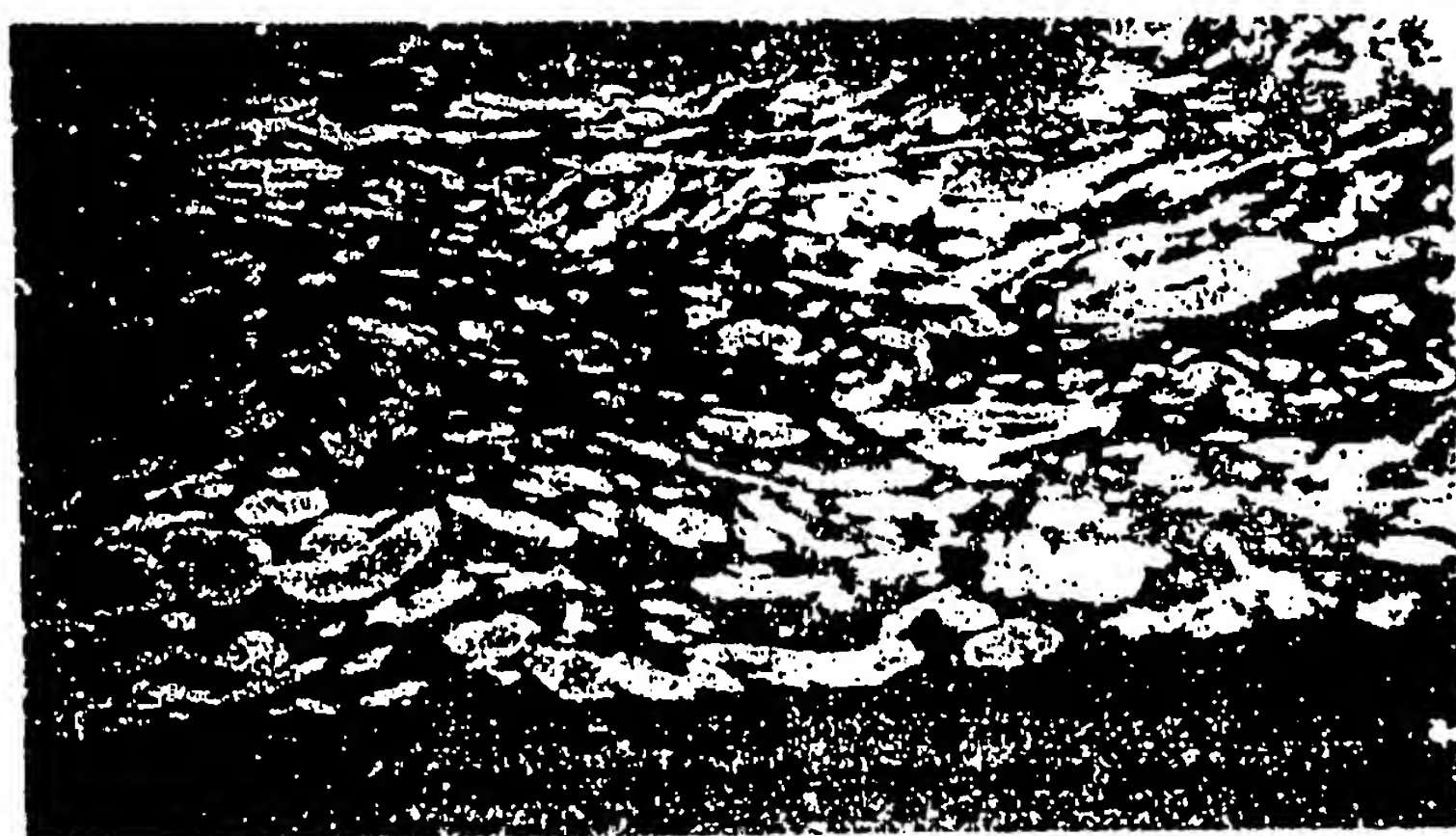


FIG. 10

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FIG. 11



FIG. 12

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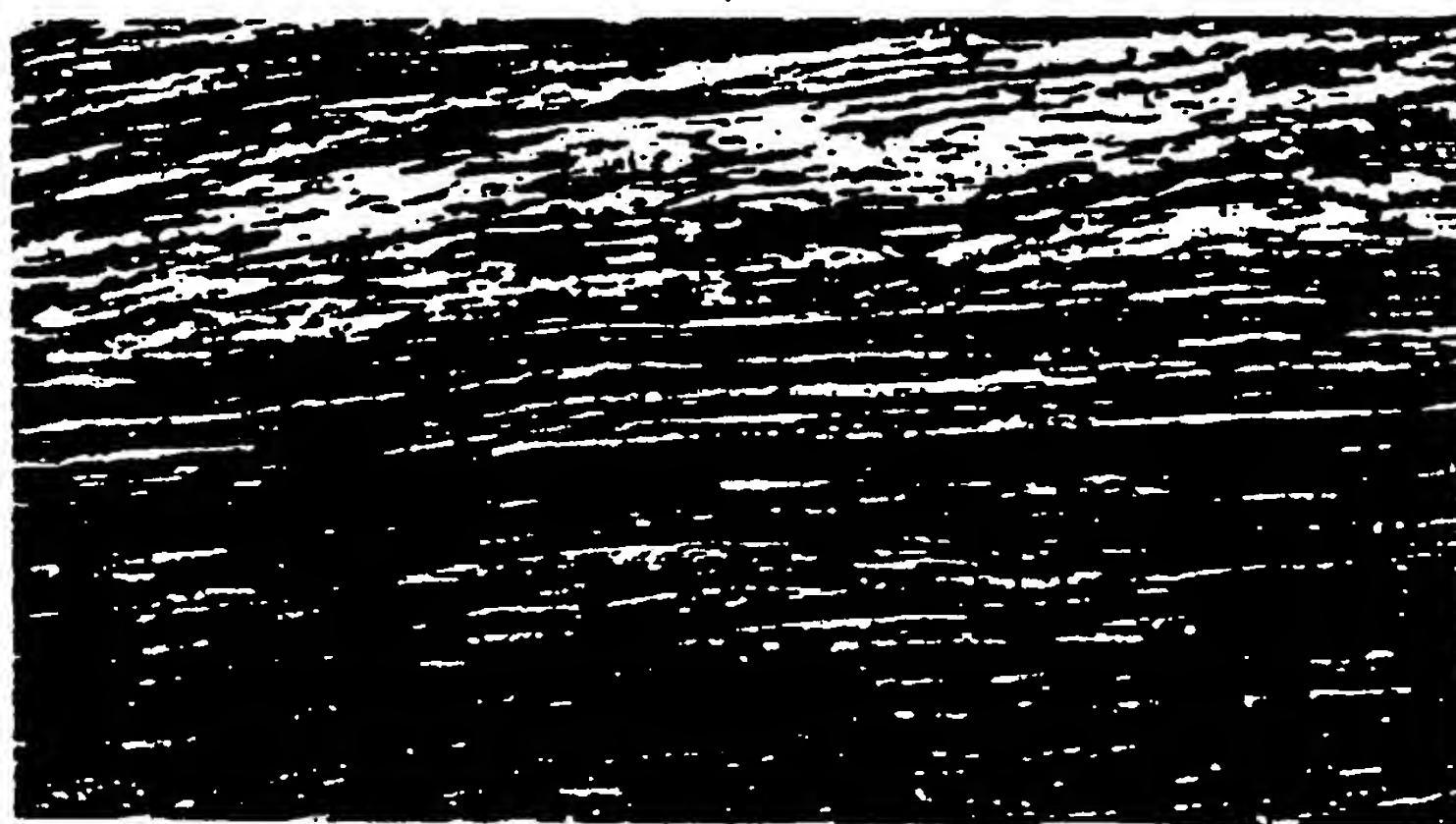


FIG. 13

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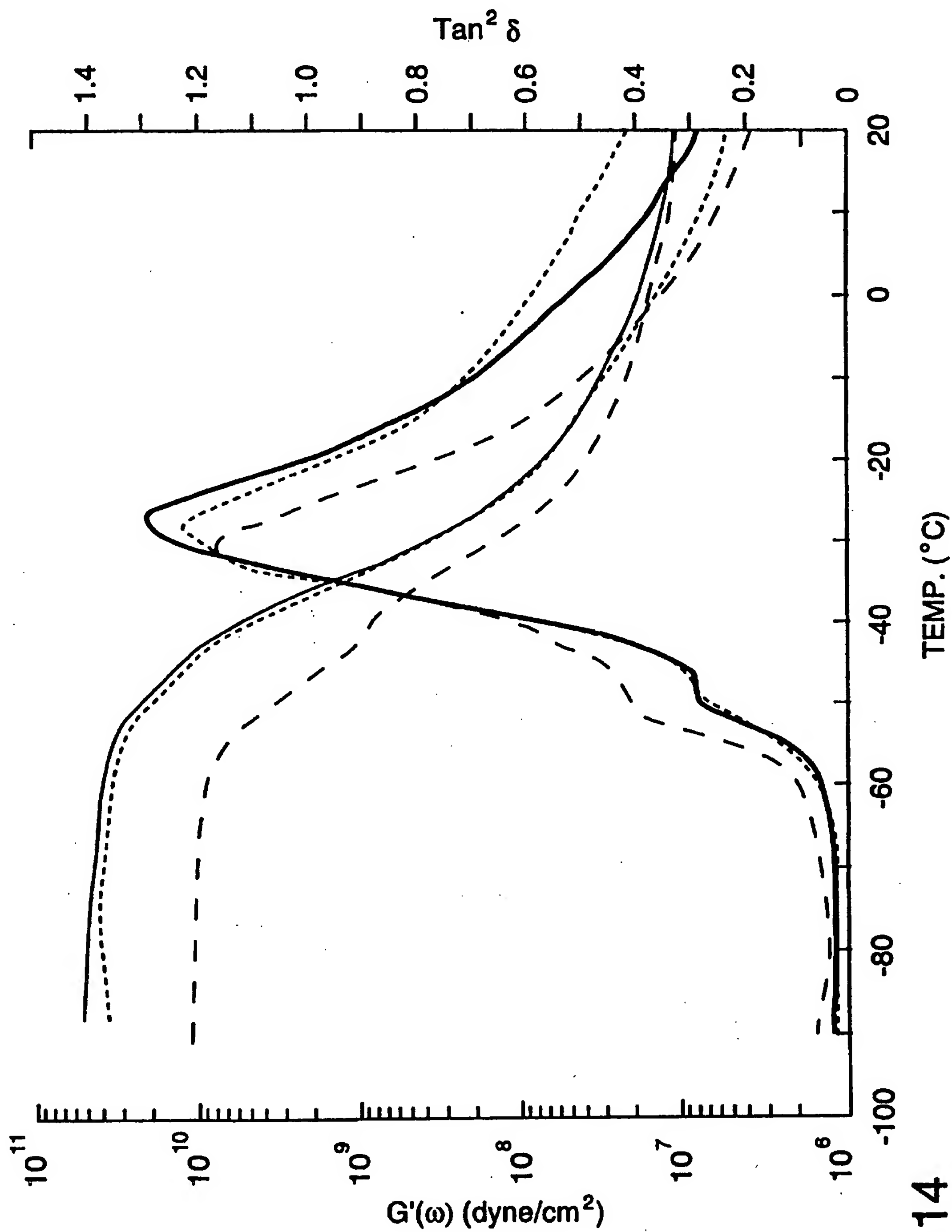


FIG.14

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 96/00430

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C09J133/08 C09J121/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 437 068 (MINNESOTA MINING & MFG) 17 July 1991 see claim 1 -----	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

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Date of the actual completion of the international search

2 May 1996

Date of mailing of the international search report

05.06.96

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl,  
Fax (+ 31-70) 340-3016

Authorized officer

Schueler, D



# INTERNATIONAL SEARCH REPORT

Information on patent family members

Int. Application No

PCT/US 96/00430

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-0437068	17-07-91	US-A- 5024880	18-06-91
		AU-B- 631951	10-12-92
		AU-B- 6800190	04-07-91
		CA-A- 2031601	04-07-91
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		ES-T- 2069032	01-05-95
		JP-A- 3294377	25-12-91
		JP-B- 7057860	21-06-95
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